MOLYBDENUM IN THE ENVIRONMENT AND IMPLICATIONS TO MINE DECOMMISSIONING IN BRITISH COLUMBIA

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ABSTRACT

An environmental concern of several mines in BC is the potential impact of elevated concentrations of molybdenum. Possible impacts of molybdenum include effects on agricultural land use where disturbed land is returned to forage production and/or grazing, and effects on the quality of water, used either as fish habitat or irrigation supply. A review of research concerning molybdenum in the environment as it relates to BC mining conditions was initiated by Environment Canada and BC Ministry of Energy Mines and Petroleum Resources. This study, funded by CANMET through the Canada-BC Agreement on Mineral Development, involved a review of the published literature and non-published results of studies carried out by BC mines and other sources. Critical to the assessment of the literature and other research results is an understanding of the soil and aquatic chemistry conditions for molybdenum bearing mine wastes in BC. In addition to a review of the presently available information on the molybdenum concern, the study addressed critical questions which remain to be answered and proposed how government and industry could resolve these issues.

INTRODUCTION

This paper outlines the findings of a recently completed report *A Review of Molybdenum in the Environment and Implications to Mine Decommissioning in British Columbia* (Jones *et al* 1994). The report presents a review of published scientific literature, and non-published studies conducted by British Columbia mines and government agencies to assess climate, soil and water conditions both in the natural state and after mining disturbances with reference to factors influencing molybdenum uptake in plants, fish, ungulates and wildlife and differences in species tolerance of molybdenum.

In British Columbia, several copper-molybdenum mining properties could potentially produce vegetation or release waters containing elevated molybdenum concentrations. The principal properties of concern are porphyry copper-molybdenum deposits, and include Brenda, Endako, and Highland Valley Copper; monitoring data and research studies conducted by these mines was provided and reviewed for this study. Limited data was available from other copper-molybdenum porphyry mines with potential molybdenum concerns including Bell, Boss Mountain, Gibraltar, Granisle, Island Copper, and Similco.

A risk assessment approach was utilized in the review to provide a framework for evaluating the potential hazards of molybdenum in the environment. The report initially provides a discussion of the potential exposure concentrations of molybdenum in water, soil, rock, plants and animals both in the natural state and following mining. The report then focuses on the characterization of the exposure settings, identification of potential receptors, routes of exposure and quantification of exposure with specific reference to domestic livestock, wildlife and aquatic life. The report concludes with an assessment of toxicity information pertaining to cattle, wildlife and fish, and discusses information necessary to quantify a risk assessment.

ASSESSMENT OF POTENTIAL EXPOSURE CONCENTRATIONS

To evaluate the potential risk posed by molybdenum in the environment the report initially identifies the potential exposure concentrations in ambient settings and mining-disturbed settings. Following sections evaluate the chemistry and concentrations of molybdenum in ambient conditions, and associated with mining activities.

Molybdate is highly soluble at higher pHs, as opposed to other metals, such as copper and uranium which are precipitated as hydroxides at high pHs. As the pH becomes more weakly acidic, molybdate polymerizes to form polyrnolybdate ions (Cotton & Wilkinson, 1972). At approximately pH 6, the first polymerized species, $Mo_7O_{24}^{6-}$, is detectable, and under slightly more acidic solutions octamolybdate, $Mo_8O_{26}^{4-}$, may be the predominant species. These isopolymolybdate ions may become hydrated or protonated, and then actively bind to counter-ions. However, under more oxidizing conditions and at pH's below 5, which are likely to be found in some industrial effluents or acid-mine drainages, bimolybdate ($HMoO_4^-$) is the predominant species. At very low pH's and under less oxidizing conditions, MoO_2^+ is the main species present (Chappell *et al.,* 1979).

It appears that the main fate processes for molybdate in water are:

- co-precipitation with iron and aluminum oxides and hydroxides
- interaction with hydrogen sulphide to form an insoluble MoS_2
- complexation with dissolved organic compounds (based on soil studies) and uptake by aquatic vegetation including algae.

Geochemistry of Molybdenum Ores and Mining Wastes

Molybdenum is mainly found in acidic and basic igneous rocks and organic-rich argillaceous sediments. Molybdenite (MoS_2) is the primary ehalcophilic molybdenum mineral, and other ores of commercial importance include molybdite (MoO_3), wulfenite ($PbMoO_4$), powellite ($CaMoO_4$) and ilsemannite (Mo_3O_8). In British Columbia, molybdenum mineralization is most frequently associated porphyry copper deposits of the calc-alkaline suite.

Spatially, the majority of the porphyry copper and porphyry copper-molybdenum deposits occur within the Intermontane Belt, comprising a northwest trending belt in the central interior region of the Canadian Cordillera. In addition, several porphyry deposits with molybdenum associations have been identified on Vancouver Island within the Insular Belt, west of the Coast Plutonic complex. Porphyry deposits are typically large tonnage, low grade deposits. The copper-molybdenum and molybdenum deposits are related to felsic, calc-alkaline intrusive bodies and host rocks range in composition from granodiorite to granite or syenite to quartz monzonite.

Molybdenum in Soils

Molybdenum concentrations and the chemistry of molybdenum in soils are important considerations in the assessment of potential exposure concentrations as molybdenum availability in soil can influence molybdenum concentrations of vegetation and forage produced on the site.

Total molybdenum concentrations in soils were reported (Highland Valley Copper, 1994) for the area to the north and east of the Highland Valley mine site. Total molybdenum content of all the soils sampled were greater than the normal range for soils. The typical range for total molybdenum in this study was 12 to 24 ppm. Higher concentrations of molybdenum occurred in sample locations within the active floodplain of drainages.

In a study conducted in the vicinity of Endako Mines (Norecol 1990) total soil molybdenum concentrations ranged from 2.3 ppm to 258 ppm. Within a drainage basin, soil molybdenum levels decreased with distance downstream from the mine site. In one drainage molybdenum levels also decreased with distance from the stream, but this trend was not apparent in the other drainages.

The following factors have been reported in the literature as significant to molybdenum chemistry in soils:

- adsorption to iron oxides and hydroxides, metahalloysite, nontronite and kaolinite, illite and allophanes, oxides of titanium and aluminum and organic matter.
- mobilization of molybdenum in soils is highly pH dependent. Overall it could be stated that as the pH of soil increases, molybdenum sorption decreases.
- Soils with reducing conditions, such as observed in poorly drained areas, may result in mobilization of MoO_4^{2-} (Allaway, 1977). Under reducing conditions, iron changes from Fe^{3+} to Fe^{2+}, forming more soluble iron molybdate compounds or complexes (Kubuta *et al.,* 1963). Additionally, the formation of soluble thiomolybdates is also said to occur under reducing conditions (Kubuta *et al.,* 1963).
- In addition to adsorption to iron oxides and hydroxides, fixation of molybdenum may occur during diagenesis of amorphous iron oxides via the formation of ferro-molybdite [$Fe_2(MoO_4)_3 \, 8H_2O$] and other crystalline forms (Barrow, 1973).
- The presence of organic matter may have varying effects on molybdenum fixation to soils.

Organic chelation of molybdenum has been observed in the presence of aerobically decomposing plant material, and the complex may then be fixed by colloidal soil organic matter over a pH range of 1.5 to 6.5 (Bloomfield and Kelso, 1973). However, under anaerobic conditions, molybdate remains relatively mobile.

Molybdenum in Plants

Molybdenum plays an important role as a micronutrient for plants in nitrogen metabolism. Plant tissue concentrations of about 0.03 - 0.15 ppm are generally adequate for plant physiological requirements; however, normal leaf tissue concentrations are in the order of about 1 ppm. Legumes have a high demand for molybdenum because it is a biochemical component of the metalloenzyme nitrogenase which fixed nitrogen in the symbiotic rhizobial nodule (Schrauer, 1977). Molybdenum concentrations vary widely between species.

High levels of molybdenum in plant tissue may result in molybdenosis (in cattle and sheep). When the ratio of copper to molybdenum in feed drops below 2:1, molybdenosis can be expected in cattle unless the copper concentration in the feed exceeds 13-16 ppm. (Committee on Medical and Biological Effects of Environmental Pollutants, 1977). The effects are discussed in Section 5.1.1 of this review. As a result, there has been a significant amount of research in the uptake of molybdenum by vegetation.

Molybdate is the dominant form of molybdenum taken up by plants from soil solution, and the soil physiochemical factors described previously will strongly affect plant tissue concentrations.

The uptake of molybdenum is correlated to the amount of molybdenum in soil solution. The availability would be dependent upon numerous complex factors. These factors include pH, soil drainage and redox potential, anions such as phophorus and sulphate, organic carbon content, and soil microflora.

Molybdenum in the Vicinity of British Columbia Mine Sites

Characterization of the molybdenum concentrations at and near British Columbia's known molybdenum producers is integral to the assessment of potential exposure concentrations. This information provides for a measure that can be compared to the baseline ambient concentrations to determine the range of potential exposure concentrations typical of each medium (water, rock, soil and plant).

Molybdenum in Water

Molybdenum concentrations in discharge and ambient water at or near several British Columbia molybdenum mining sites was reviewed. The data is limited with respect to total molybdenum but indicates a range in dissolved molybdenum concentrations of 0.003 to 0.22 mg/l in background waters, and a range of 0.005 to 11.4 mg/l reported for sites downstream of mine discharge. Data provided by Endako Mines reports the highest molybdenum concentrations both upstream and downstream of mine discharge. The pH of sampled waters range from 6.7 to 8.3 and no strong correlation is evident between molybdenum concentration and pH of sampled waters.

Soil Conditions at Mine Sites

Soils at mine sites may be comprised of previously stripped soils, overburden materials or mine wastes. Relatively few studies have been conducted on mine soils at mineral mines in British Columbia. Of particular interest to this review, are studies which correlate soil conditions with plant uptake of metals.

In a greenhouse study, grass and legume concentrations of molybdenum were determined for various overburden materials from the Valley Pit at Highland Valley Copper (Highland Valley Copper 1994). Evaluation of the range of molybdenum concentrations in overburden and vegetation indicate that there is significant variability. Overburden soil materials ranged from 1.2 to 139 ppm total molybdenum, grass concentrations ranged from 2 to 341 ppm molybdenum, and legume concentrations ranged from 2 to 1030 ppm molybdenum. Regression analysis indicated some relationship between molybdenum in grasses and total molybdenum in overburden (r^2=0.4) and between molybdenum in legumes and total molybdenum in overburden (r^2=0.5) (Highland Valley Copper, 1994).

Vegetation at Mine Sites

Molybdenum uptake by plants appears to be species specific, with legumes generally accumulating more molybdenum than grasses when grown in the same soil (Allaway 1977). Various studies at British Columbia mine sites confirm the species specificity in molybdenum uptake (Gould Gizikoff 1990; Hackinen 1986; Highland Valley Copper 1992, 1993, 1994; Jones and Associates 1993).

Studies at Highland Valley Copper and Brenda Mines illustrated very high variability in molybdenum content in vegetation within sites. The coefficient of variation for molybdenum was extremely large in comparison to copper for both legumes and grasses. The distribution of molybdenum in all plant samples is markedly more clumped (ie not a random distribution) than is copper (Highland Valley Copper, 1993; Jones and Associates 1994).

CHARACTERIZATION OF EXPOSURE SETTING AND OF POTENTIAL RECEPTORS

To determine those species which will be the potential receptors of molybdenum contained in water, soil and vegetation on mine sites, it was necessary to review the proposed land uses and reclamation products of the various molybdenum producers in British Columbia.

Reclamation plans and reports for various molybdenum producers in British Columbia were reviewed to determine proposed land uses. The most commonly listed potential receptors of elevated concentrations of molybdenum in vegetation and water in British Columbia are:

- deer
- moose
- cattle
- fish

ROUTES OF EXPOSURE TO POTENTIAL RECEPTORS AND QUANTIFICATION OF EXPOSURE

To determine the exposure of the potential receptors to molybdenum it is necessary to understand the routes by which molybdenum can enter their bodies and to quantify this exposure.

Cattle, the primary livestock receptor, can be exposed to molybdenum through ingestion of grazed forage, cut forage, soil particles and water. Beef cattle grazing in the area of the molybdenum producers is typically restricted to summer and early fall grazing. The cattle winter on pastures at lower elevations where they are typically fed hay or silage and possibly some grain supplements.

Deer and moose, the two primary wildlife receptor species, can be exposed to molybdenum through ingestion of grazed forages, trees and shrub browse, soil particles and water. The deer's diet depends on what is seasonably available. The diet of moose varies considerably but is characterized in general by use of early successional woody browse. Moose adapt to a variety of available forage, during the summer, moose utilize forbs and aquatic plants (Schmidt and Gilbert 1978).

The primary aquatic receptors are fish; the species which has been most studied at British Columbia mines is Rainbow trout. Freshwater fish can be exposed to molybdenum through uptake from food or through the gills. It is believed that the primary made of action is through the gills, since the bioconcentration factors observed with molybdenum are relatively low and this generally results from bioaccumulation from the water, not biomagnification through food consumption.

ASSESSMENT OF TOXICITY INFORMATION

A principle goal of this review of molybdenum research was the compilation and evaluation of the current body of knowledge pertaining to the toxicology of molybdenum with respect to domestic livestock, wildlife and aquatic life.

Domestic Livestock

The effects of molybdenum on domestic livestock are of great interest as decommissioning of mines may yield forage areas not previously available, but these forages may contain elevated concentrations of molybdenum.

The importance of molybdenum in ruminant nutrition dates from 1938, when it was discovered that "teart" disease in cattle was caused by excessive absorption of molybdenum. A paper by Anke and Groppel (1987) summarizes the early findings. This disease is characterized by severe scouring and loss of condition, and a secondary copper deficiency. Copper and sulphur levels in the diet were found to be of great importance. Inorganic molybdenum compounds, in the presence of sulphide, are converted to thiomolybdates in the rumen, which in turn bind copper, making it almost completely unavailable. Typical "teart" pastures have molybdenum levels of 20 - 100 ppm. Adjacent vegetation not associated with the "teart" conditions contains less than 3 - 5 ppm molybdenum.

Excessive molybdenum intakes in ruminants have been found to cause, in addition to some abnormalities and other symptoms of copper deficiency, adverse effects on reproduction. Effects noted include:

- infertility in heifers and cows;
- inhibition of oestrogen and androgen receptors *in vitro;*
- lack of libido; and,
- damage to interstitial cells and germinal epithelium.

Mills and Davis (1987) have noted that some of the effects of high levels of molybdenum are only partially eliminated by increasing the copper intake, and that some of the histopathological features of molybdenum toxicity do not relate to copper deficiency. Features suggested as characteristic of molybdenum excess *per se* include mandibular exostoses, testicular damage in rats and cattle, and disorders of phosphorus metabolism in cattle that produce skeletal abnormalities and osteoporosis.

It has been well established that a molybdenum induced copper deficiency in ruminants involves interaction between copper, molybdenum and sulphur, and the formation of thiomolybdates.

Suttle (1991) has summarized the treatment of molybdenum induced disorders in ruminants. These are animals showing characteristic hair, fleece and bone abnormalities, and impaired growth. Chronically copper deficient animals will respond to dietary copper supplements, oral boluses or injections. If thiomolybdates, as suspected, have an adverse affect on the intestinal tract mucosa, a continuous dietary supply of copper is best. If thiomolybdates have widespread and toxic effects on reproduction, it is questionable whether either oral or parental copper will be immediately effective. Alternatives such as limiting molybdenum intake would have to be considered.

Phillippo *et al.* (1987) showed very clearly that the animals supplemented with molybdenum came into oestrous later and had lower conception rates than control animals.

Wildlife

Some important wildlife species in British Columbia are ruminants, including deer, moose, and elk. It is recognized that there are differences between species in their sensitivity to copper and molybdenum. Deer are reportedly less susceptible than domestic ruminants. Ward and Nagy

(1976) fed mule deer sodium molybdate at levels ranging from 50 - 7500 ppm molybdenum. The deer were slower to develop symptoms than cattle would have been at these levels. However, the deer reduced their feed intakes at the higher levels of molybdenum, and lost weight. Four of the seven animals eventually died. The diet used in the experiment was a concentrate pellet, markedly different from the natural diet of mule deer.

Fish

Much of the available laboratory test results indicate that fish can tolerate exposure to relatively high levels of molybdenum (Eisler, 1989). The acute median lethal concentrations (96-hour LC50's) of molybdenum (from sodium molybdate) to rainbow trout were 800 mg/f for fish 20 mm in length, and 1,320 mg/l for fish 55 mm in length (McConnell, 1977). As part of this same study, it was found that a one-year exposure to molybdenum did not exert a toxic effect on eyed eggs, sac-fry (alevins), or fingerling stages of rainbow trout at concentrations up to 17 mg/l. In another more recent study (Hamilton and Buhl 1990), the 96-hour LC50's of eyed eggs, alevins, and fry of both coho and Chinook salmon were >1000 mg/l.

In contrast to these findings, when rainbow trout were exposed to molybdenum immediately after egg fertilization until 4 days post-hatch (28 days), the chronic threshold-effect concentration was 0.73 mg/l on one occasion (Birge, 1978) and 0.79 mg/l in a later study (Birge *et al.,* 1980). However, a recent study, in which coho salmon were exposed to molybdenum 30 minutes after fertilization until 20 days post-hatch (20 weeks), found no significant effects in molybdenum at concentrations =;15 mg/l (Ennover, manuscript in preparation). Differences in test species, test temperature, test apparatus, and dilution water likely contributed to these diverse findings, and make it difficult to compare the results of the Einnover and Birge studies.

Both laboratory and field studies have suggested that fish are able to regulate molybdenum accumulation when ambient molybdenum levels are elevated in test solutions or receiving waters. Ward (1973) found that concentrations of molybdenum in tissues of rainbow trout, sampled from receiving waters, increased only slightly with increase in molybdenum concentration of the water, and suggested that there might be a threshold beyond which excess molybdenum is efficiently excreted. He also noted that rainbow trout exhibited generally higher tissue concentrations of molybdenum than kokanee salmon, sampled from the same receiving waters.

Fisheries studies and *in situ* exposures of fish in receiving waters of British Columbia mines appear to corroborate findings by other researchers which indicate that molybdenum is not very toxic to fish, nor does it biomagnify in their tissues (Mathieu 1994 manuscript in preparation; Hatfield Consultants Ltd. 1992 and 1993; Patterson, personal communication 1994).

INFORMATION NECESSARY TO COMPLETE A RISK ASSESSMENT

To complete a risk assessment for molybdenum on the target receptors, cattle, deer, moose and fish, additional information will be required. The report addresses the limitation within the existing data base and suggest research requirements.

Cattle

It is clear from the review of the literature that excessive levels of molybdenum in forages can create severe problems in livestock. Some, but not all of these problems can be alleviated by ensuring adequate copper status in the animals. There is evidence, however, that other disorders, attributable to molybdenum *per se,* and independent of copper status exist. The possibility of impaired reproduction is of particular concern.

In British Columbia we have essentially no useful information regarding the longterm effects of consuming the high molybdenum forages which are associated with molybdenum-copper mining. This observation applies to both domestic and wild ruminants, and the concern about reproduction is of equal importance to both. Any approach taken to providing new information should include, as a high priority, consideration of reproduction.

An experimental approach was suggested to consider possible reproductive effects and other potential serious metabolic effects.

Wildlife

As discussed for cattle, the information necessary to determine the longterm risks to wildlife of consuming the high molybdenum forages and browse associated with molybdenum mining in British Columbia is not presently available.

Monitoring studies are suggested for the important wildlife species, deer and moose. These programs require considerable research effort to determine use of mine areas by animals through radio equipped tracking collars.

Aquatic Life

This review indicates that fish can tolerate exposure to relatively high levels of molybdenum. However, early life stages of fish might, under certain test conditions using the ionic species of this metal, be vulnerable to molybdenum toxicity at concentrations that could occur within receiving waters. Results from presently available studies are conflicting and inconclusive. Therefore, to adequately assess the potential risk to fish populations due to their acute or chronic exposure to molybdenum in receiving waters, we recommend that early life-stage tests are undertaken according to Environment Canada (1992) with sodium molybdate, and dilutions of mine effluent, using both local receiving waters and laboratory water as the dilution water.

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