

## SHOULD THE EFFLUENT LIMITS FOR MANGANESE BE MODIFIED?<sup>1</sup>

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**Abstract.**--As a result of the 1972 and 1977 Clean Water Acts, effluent mine water is limited to a 30-day average concentration of 2 mg/L manganese and a single-day maximum of 4 mg/L. These limits were selected by EPA after a survey of mine water treatment facilities indicated that these values could be achieved consistently, and that in the process of removing manganese, other trace metals were also controlled. There are, however, several reasonable arguments against such low limits. Current mining operations often face much higher levels of manganese than were considered by EPA when they established the limits. As a result, many operators must raise the pH of mine water to 10 or above, increasing their chemical treatment costs by as much as 100 pct. Precipitation of manganese then lowers the pH somewhat, but an effluent pH of 9 or even higher is common. Can the limits be safely eased? Based on recent studies, it appears that manganese is only toxic to fish at low concentrations when the stream water is exceptionally soft and pure. Except in such rare instances, manganese can be considered to be about as toxic as potassium. Our work also indicates that the other, trace metals of concern are removed at a pH below 9. It is therefore suggested that the manganese effluent limits should be reexamined in light of this information.

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### INTRODUCTION

Regulations, once implemented, are rarely reexamined by the enforcement agency unless such action is dictated by legislation. The threat of court action by environmental groups also acts to inhibit relaxation of regulations. It is therefore the responsibility of the affected group to

periodically reexamine its regulatory limits and seek modification or exception to regulations, where appropriate. Eastern coal mine operators have repeatedly called for a less stringent limit on manganese, based on their difficulty in meeting the effluent limits, the relatively high cost associated with doing so, and their perception that the regulation serves little purpose in protecting aquatic life and downstream consumers.

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It is the intent of this paper to explain the rationale for the Federal effluent limits on manganese, to reexamine those limits based on subsequent studies, and to compare the costs and associated benefits of current regulations with more lenient alternatives.

### THE REGULATION OF MANGANESE

Regulation of manganese was a result of the 1972 and 1977 Clean Water Acts, which directed EPA to establish regulations that were technology-

driven. Specifically the 1972 Act required regulations based on the "best practicable control technology" (BPT). In response, EPA promulgated new source performance standards for the mining industry in the U.S. Federal Register in 1976. The 1977 Act required regulations based on the potentially more rigorous "best achievable control technology" (BAT), citing 129 toxic substances, including 13 metals, that were termed "priority pollutants." Manganese was initially included on this list due to toxicity concerns (discussed later) and the discoloration problems manganese can cause to downstream water users if supplies are contaminated at concentrations of 0.2 mg/L (reviewed in Kleinmann and Watzlaf 1986.)

As a result of a series of surveys and site visits, EPA initially established a BPT effluent limit for manganese of 4 mg/L maximum (daily) and a 30-day average limit of 2 mg/L (U.S. Federal Register 1976). The BAT regulations that followed left these limits in place. However, although the principal motivation for regulating manganese initially was its potential role as a pollutant, the impetus changed during EPA's regulatory development process. EPA concluded that manganese toxicity was not a serious problem. Downstream use was still a concern, but more important was the finding that by regulating manganese, EPA could avoid imposing regulatory limits on eight other more toxic metals (arsenic, chromium, copper, lead, mercury, nickel, selenium and zinc) that were found to be present occasionally at relatively low concentrations (Weideman 1982). Manganese was selected as a surrogate for these metals for two reasons. First, when the other priority pollutants were present, the more-common manganese was also found. Second, when manganese was removed during treatment in a conventional treatment plant by addition of alkalinity, these other metals of concern were also precipitated.

Why were the manganese limits set at 2 and 4 mg/L, as opposed to some other values? EPA, in its survey found that where acid mine water treatment plants were operating properly, manganese could be reduced to about 2 mg/L. State permits, and other effluent limits on point source discharges now typically mirror these BAT limits (cf. OSMRE program performance standards--U.S. Code of Federal Regulations 1985, Nalesnik Associates 1980).

However, the average manganese concentrations of the untreated mine waters in the survey used to develop the BAT guidelines were only 4.9 mg/L at underground mines and 17.7 mg/L at surface mines (Weideman 1982). Manganese did not exceed 63 mg/L at any surveyed acid mine water site. Our work indicates that there are today many mine sites with manganese levels much higher than those found by EPA a decade ago. Also, the average iron-manganese ratio in the EPA study was 2.6 for surface mines and 27.6 for the surveyed underground mines with acid water problems (ibid). Table 1 lists some examples of high manganese values, selected from analyses of various samples collected by the Bureau of Mines during the past few years. At many of these sites, concentrations of manganese were actually higher than those of iron. Recent laboratory tests and previous work have shown that removal of manganese becomes increasingly difficult as the iron-manganese ratio falls below about 2.5, presumably owing to the decrease of co-precipitation of manganese on ferric hydroxide (Ackman and Erickson 1986).

Table 1.- Manganese and iron concentrations in acid mine water, from analyses on record in authors' files.

Site	Total Mn (mg/L)	Total Fe (mg/L)	Fe/Mn
PA - Greene Co.	211.4	729.4	3.5
PA - Greene Co.	121.0	286.9	2.4
PA - Venango Co.	87.7	51.3	0.6
PA - Clarion Co.	91.2	31.6	0.3
PA - Clarion Co.	106.8	691.6	6.5
PA - Clarion Co.	40.7	20.5	0.5
PA - Centre Co.	130.6	193.2	1.5
PA - Clearfield Co.	30.6	50.6	1.7
PA - Clearfield Co.	138.3	116.7	0.8
PA - Clearfield Co.	160.6	131.7	0.8
PA - Clearfield Co.	122.2	13.3	0.1
PA - Clearfield Co.	82.7	39.7	0.5
PA - Clearfield Co.	116.5	106.6	0.9
PA - Clearfield Co.	102.8	42.9	0.4
PA - Westmoreland Co.	138.0	100.0	0.7
PA - Westmoreland Co.	202.4	680.8	3.4
PA - Westmoreland Co.	108.0	19.7	0.2
PA - Westmoreland Co.	79.0	38.2	0.5
WV - Upshur Co.	158.6	43.9	0.3
WV - Upshur Co.	180.0	90.6	0.5
WV - Upshur Co.	69.8	21.3	0.3
WV - Kanawha Co.	257.0	147.0	0.6
KY - Pike Co.	48.0	102.5	2.1

Thus, EPA's decision reflects the relatively high iron-manganese ratios and the relatively low concentrations of manganese that were observed in its study. The discharge criteria were not selected to satisfy perceived downstream requirements nor because such low limits were necessary for trace metal removal; they were targeted at the level observed to occur in the effluent waters of the surveyed water treatment facilities. If the sites listed in table 1 had been included in the EPA survey, it is quite possible that a higher discharge standard would have been selected, due to the difficulty of meeting such stringent limits at these sites.

#### THE COST OF MANGANESE REMOVAL

Cost is also a factor in determining BAT limits. If manganese must be removed by increasing the pH to 10 or above, the material costs of chemical treatment are increased by as much as 100 pct over the costs of iron removal. The alternative technique of treating to a neutral pH, aerating to remove the iron, and then using a chemical oxidant to remove the manganese increases costs by an additional 200-300 pct (Watzlaf 1985, Kleinmann et al. 1985) and fails to remove some of the other trace metals (Watzlaf 1988).

Figure 1 illustrates the costs of removing manganese by adding sodium hydroxide with optimal control of pH, as calculated for three mine sites with manganese concentrations of 12 to 100 mg/L. To meet the current 2mg/L limit, a site operator facing a typical influent manganese concentration of 25 mg/L would spend at least \$300 extra for chemicals to treat a million gallons of water beyond the pH of 8 normally needed to remove iron. At a standard of 5 mg/L, this cost is reduced by \$100. If the limit were 10 mg/L, chemical costs would be reduced another \$100 for every million gallons of water. So, for a treatment facility handling 1 million gal/day, the annual savings in chemical costs alone would be at least \$71,200 if the effluent limit was raised from 2 mg/L to 10 mg/L and half that if the effluent limit was raised to 5 mg/L.

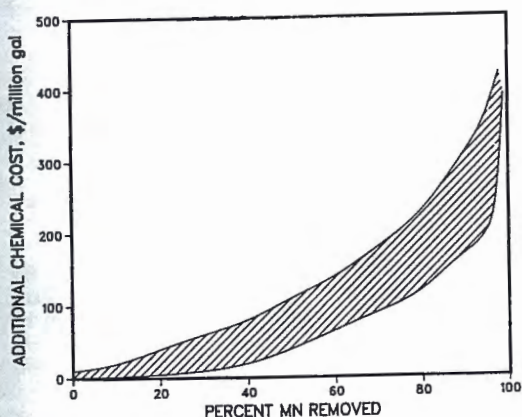


Figure 1.--Range of additional chemical costs (sodium hydroxide) for manganese removal. Chemical cost for pH adjustment to achieve regulatory compliance with iron standard is not included. Graph is based on results from three mine sites where manganese concentration ranged from 12 to 100 mg/L.

Other costs would also decrease if effluent limits for manganese were made less stringent, since increasing the pH above 9 greatly increases sedimentation basin and sludge disposal requirements. Nicholas and Foree (1979) calculated that a site that utilized sodium hydroxide to neutralize mine water that contained 74 mg/L total iron and 26 mg/L total manganese, would require 31 pct more basin area as the required pH was increased from 9 to 10, even though the amount of additional manganese removed was minimal. At another site with 4 times more iron and manganese, increasing the pH with lime from 8.7 to 9.75 increased the basin area requirements from 8,000 ft<sup>2</sup> to 12,857 ft<sup>2</sup>, with an associated decrease in discharge water manganese from 3.5 mg/L to 0.2 mg/L (ibid).

Many mine operators report that fluctuations in mine water quality and quantity cause additional mine water treatment problems. This is especially true at sites where the influent iron/manganese ratio is less than 2.5, since these are the sites where the operators must raise the pH to 10.0 or higher to satisfy the existing manganese criteria. Normal fluctuations can increase or decrease the amount of alkalinity required; if the operator fails to adjust the flow correctly, the effluent water is out of compliance due to either manganese or an excessively high pH, even after precipitation of the metals brings the pH down. Also, excessively high pH treatment can cause precipitated aluminum and iron to redissolve. Water treatment thus becomes a balancing act, with potential noncompliance (and fines) an everyday occurrence.

It should also be remembered that the expense of water treatment continues long after mining has ceased and coal sales are no longer producing revenue. There are now several reclaimed mine sites where the mine water meets all of the water quality criteria except for manganese. In such cases, water treatment has had to be maintained to control manganese, typically without release of bond money. For example, one otherwise-legal discharge in Somerset County, PA, with 8 to 10 mg/L of manganese is causing a company to spend \$52,000 a year on sodium carbonate (soda ash briquettes), while at the same time preventing the release of \$75,000 in bond money. At another site in the same county, a \$200,000 bond is being held due to manganese in the mine discharge water.

#### ENVIRONMENTAL CONCERNS

Before effluent limits for manganese can be modified, one must reexamine the environmental concerns that led to manganese being regulated. Since manganese is serving as a surrogate for other more toxic metals, that aspect will be addressed first. Second, the issue of fish toxicity will be discussed in some detail. Finally, there is the question of whether any downstream water supplies would be adversely affected. This is a function of site-specific dilution and existing levels of contamination, but laws that regulate manganese in public water supplies and streams would come into play if there was a problem (reviewed by Nalesnik Associates 1980).



## Trace Metal Removal

A study by EPA (reported in Weideman 1982 and Going 1980) tested removal of spiked levels of the 13 metals on the priority pollutant list, including the 8 metals listed earlier. The metals of concern were all reduced to 0.07 mg/L or less at pH 8.4. Tests were also conducted in our laboratory using two mine waters with very different iron-manganese ratios, and naturally high levels of nickel, zinc, copper, and chromium. For both mine waters, zinc, copper, and chromium concentrations were reduced to less than 0.05 mg/L at pH 8.0, 7.0, and 6.8, respectively. Nickel was reduced to below 0.05 mg/L at pH 8.0 in the high iron mine water and at pH 8.8 in the low iron mine water. By way of comparison, metal finishing plants must meet BAT effluent limits of 1.5 - 2.4 mg/L for these metals. In both mine waters, reducing the concentration of manganese to 20 mg/L was sufficient to reduce all of these trace metals to 0.1 mg/L or less. This experiment is described in more detail elsewhere in this volume (Watzlaf 1988). Since a more lenient manganese limit (for example, 10 mg/L) would still require treatment to a pH of 8.5 - 9.0, it would appear that a standard of 2 mg/L for manganese is not required to assure removal of other trace metals.

## Toxicity of Manganese to Aquatic Organisms

Regulators selecting a manganese standard for the mining industry found that published literature on the toxicity of manganese to aquatic organisms was confusing and apparently contradictory, with reported toxic values ranging from 1.5 mg/L to 3,400 mg/L. This wide range can be attributed to the different species of fish used in the various studies and to differences in experimental conditions. As a case in point, let us critically examine the oldest often-cited study. In 1915, Thomas reported on the effect of various substances on *Fundulus heteroclitus*, a small minnow. In its natural environment (salt water), the minnow tolerated manganese at all levels tested (not clearly specified, but probably up to 200 mg/L  $MnCl_2$ ). Thomas observed, however, that the minnow could survive in brackish water, and through experimentation, determined that it could even survive in fresh water. Thomas repeated his experiments in tap water simply to see what effect the fresh water would have, and reported that 12 mg/L  $MnCl_2$  killed the minnows in 6 days. Subsequent publications cite the Thomas study by reporting this toxicity value and therefore are misleading (cf. Hill 1972, Weideman 1982).

Most of the reported studies examined the toxicity of manganese in tap water. Jones (1939) found 50 mg/L manganese to be toxic to sticklebacks, while Kaemmerer and Erichsen (1951) reported that 50 mg/L was tolerated, under similar conditions. Oshima (1953) experimented with freshwater eels and reported no deaths in 50 hours of exposure to manganese concentrations greater than 2,700 mg/L. Iwao (1936), working with

freshwater cyprinodonts (*Orizias latipes*), found 3,400 mg/L manganese to be the 24-hour toxic lethal limit. Clemens and Sneed (1959) reported that channel catfish fingerlings tolerated a manganese disodium EDTA solution for over 96 hours at concentrations greater than 500 mg/L (equivalent to 40 mg/L Mn). In tap water that contains 120 mg/L hardness,\* Agrawal and Srivastava (1980) determined a 96-hour  $LC_{50}$ \*\* for *Colisa fasciatus* to be 2,850 mg/L.

Trout, especially rainbow trout, are very sensitive to manganese. Lewis (1976), using distilled water, observed that the mortality of rainbow trout eggs increased from 7 pct at 0 mg/L of manganese to 12 pct at 1 mg/L, to 22 pct at 5 mg/L, and to 30 pct at 10 mg/L. Fry and adult rainbow trout were unaffected by 10 mg/L or less. England (1977), using lake water with very low hardness (2 mg/L), determined a 96-hour  $LC_{50}$  for manganese to be 24.7 mg/L. England and Cumming (1971) determined that the tolerance limit of rainbow trout fingerlings in 96-hour laboratory tests was 16 mg/L manganese for 59 to 65 mg/L hardness. Hill (1972), working with water that had a total hardness of 120 mg/L, found that the tolerance limit was 50 mg/L  $Mn^{2+}$  for juvenile rainbow trout and 88 mg/L<sup>2+</sup> for adult rainbow trout, but that these tolerance limits decreased in silty water.

Much lower tolerance limits were reported in a series of papers that followed the deaths of rainbow trout at soft water (5 mg/L hardness) fish hatcheries in the Chattahoochee River watershed in Georgia and Arkansas. Although not previously considered by EPA in its regulatory decisions, these studies, when taken together, actually clarify much of the apparent contradiction found in the studies already described. Ingols (1976) found that 1.0 mg/L manganese caused the death of rainbow trout. Ogelsby et al. (1978), suggested that humic substances might also have played a role in the death of rainbow and brown trout, but this was ruled out by Grizzle (1981), who experimented with rainbow trout, brown trout, brook trout, and yellow perch in the vicinity of Buford Dam, Georgia, and found that only the first two were sensitive to low levels of manganese. This was followed by a series of bioassay experiments at Buford Dam and the associated Lake Sidney Lanier by Lehman et al. (1982) that demonstrated that suspended manganese was not significant but that the 48-hour  $LC_{50}$  for  $Mn^{2+}$  was approximately 0.65 mg/L (interestingly,  $Fe^{2+}$  showed almost identical toxicity).

It should be stressed that these unusually low tolerances have all been associated with one watershed that contains very low levels of hardness. Moreover, Lehman et al. (ibid) found that adding 10 mg/L of hardness prevented trout mortality at 1.0 mg/L  $Mn^{2+}$ . Even more important, when 100 mg/L calcium (250 mg/L hardness) was added, manganese was not harmful to rainbow trout at a concentration of 24 mg/L over a 20-hr period (Ingols 1976). The results of Hill (1972), England and Cumming (1971), Lewis (1976) and England (1977), discussed earlier, can be seen to also support the concept that hardness protects the fish from manganese.

Regarding other aquatic organisms, Lewis (1978) determined a 96-hour  $LC_{50}$  for juvenile longfin dace to be 130 mg/L manganese in water with

\*Throughout this paper, hardness is expressed as mg/L  $CaCO_3$ .

\*\* $LC_{50}$  refers to the lethal concentration for 50 pct of the test population.

224 mg/L hardness. The flatworm can tolerate up to 700 mg/L (as  $MnCl_2$ ) or 660 mg/L (as  $Mn(NO_3)_2$ ) (Jones 1940). The threshold limit for *Daphnia magna* was 50 mg/L (as  $MnCl_2$ ) (Bringmann and Kuhn 1959). Finally, a 7-day test at 15 mg/L of manganese had no adverse effects on crustacea, worms, and insect larvae (Schweiger 1957). In general, several investigators have concluded that manganese is only slightly more toxic to aquatic organisms than potassium (Doudoroff and Katz 1953, Jones 1939).

#### CONCLUSION AND DISCUSSION

Manganese can have an adverse effect on downstream water users and certain fish. In addition, the use of manganese as a surrogate for toxic metals at conventional mine water treatment facilities is a sensible alternative to regulating a long list of metals. However, the specific industry-wide effluent limits that were adopted, based primarily on observed levels of manganese attained at certain treatment plants, appear to be more stringent than required or intended. The range of water quality being treated and the treatment techniques being used have changed significantly since the EPA survey a decade ago. At the same time, more recent information that relates manganese toxicity to fish to hardness clarifies much of the confusion that previously existed in that literature. In addition, it would appear that even high levels of toxic metals are reduced to acceptable limits in treatment of mine water to manganese concentrations of 10 to 20 mg/L. Also, occasional analysis for other metals would now be a preferable alternative to treating to a high pH, although to be fair, a decade ago this option might have been difficult for the mining industry due to a lack of laboratory facilities.

A possible interim measure is a case-by-case adjustment of effluent limits. Specifically, the U.S. Code of Federal Regulations, Title 40, Chapter 1, Part 125 (1985), states that:

(b) In establishing national limits, EPA takes into account all the information it can collect, develop and solicit regarding the factors listed in sections 304(b) and 304(g) of the Act. In some cases, however, data which could affect these national limits as they apply to a particular discharge may not be available or may not be considered during their development. As a result, it may be necessary on a case-by-case basis to adjust the national limits, and make them either more or less stringent as they apply to certain dischargers within an industrial category or subcategory. This will only be done if data specific to that discharge indicates it presents factors fundamentally different from those considered by EPA in developing the limit at issue.

Perhaps owing to the complications caused by the superimposition of OSMRE and EPA enforcement, no such exemption has ever been granted for a site with manganese problems. However, allowing such flexibility for manganese would appear to be reasonable, if the site was otherwise suitable. Specifically, if the trace metals of concern are either absent or removed at pH 9 or below, if the stream water has sufficient hardness to protect sensitive fish, and if sufficient dilution is

available to avoid adverse effects on downstream users, then it would appear that more lenient effluent limits could be substituted without adverse consequences. Also, under current regulations, more lenient effluent limits for surface mines can be negotiated with the State regulatory agency after mining is completed, since the Federal effluent limits for manganese are not applicable (Weideman 1982). At sites where manganese is the only water quality problem, a favorable analysis with respect to such factors as hardness, downstream water users, etc., should result in a higher negotiated limit.

To conclude, the effluent limits for manganese were selected based on the best information available at the time; it would now appear to be a suitable time to reexamine those limits to determine if they can be made more lenient without harm to the environment.

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