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20 Years of Mercury Re-emission – What Do We Know?

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ABSTRACT

While conducting fundamental research in the 1990s, Babcock & Wilcox (B&W) observed a wide range of mercury emission collection rates across the wet flue gas desulfurization (FGD) systems based on the type of particulate collector which was positioned upstream of the gas flow. The term ‘mercury re-emission’ was used to describe the phenomenon of a higher concentration of elemental mercury exiting the absorber when compared to what was entering the absorber. This paper will review the experience and knowledge gained as the industry more clearly understands what is necessary for achieving Mercury and Air Toxics Standards (MATS) limits when a wet FGD system is part of the installed air quality control system.

Results from several installations will show current costs of compliance, along with the various operational strategies to utilize a wet FGD for mercury control.

INTRODUCTION

Mercury (Atomic Symbol: Hg) can enter the environment from a number of natural and man-made sources, including coal combustion for power generation {Pavlish et al 1}. In the United States (U.S.), the Environmental Protection Agency (EPA) requires electric generating units (EGUs) firing bituminous and subbituminous coals to achieve stack mercury emissions at or below 1.2 lb/TBtu. For units firing lignite coal the limit is 4.0 lb/TBtu as per the Mercury and Air Toxics Standards (MATS) rule. As of April 2016, all EGUs must comply with MATS. To achieve MATS compliance, technology choices made by EGUs to achieve the necessary Hg reductions include coal additives, sorbent injection additives via flue injections, wet FGD additives, and wet FGD sorbents to name a few {Feeney et al 2}. The amount of Hg in coal varies significantly by the region and can be as high as 0.24 ppm for the Northern Appalachian coal to about 0.08 ppm for Powder River Basin (PRB) coal.

During the coal combustion process, all of the mercury is released in the elemental (Hg^0) form. As the flue gas cools after passing through the economizer section of the furnace, some of the elemental Hg gets oxidized by the native halogen species, such as HCl in the flue gas. Some of the oxidized mercury gets adsorbed on the unburned carbon in the ash which is also present in the flue gas. This mercury is then removed by the particulate collection devices such as electrostatic precipitators (ESP) or fabric filter (FF). Remaining oxidized mercury can either be removed by other air quality control system (AQCS) equipment such as flue gas desulfurization (FGD) systems or it can be released into the atmosphere along with the flue gas from the stack

{Chothani et al 3}. Wet FGD systems are normally located downstream of the particulate collection device. Due to the high removal efficiency, reasonable cost, and wide applicability, the lime/limestone wet scrubber has become the preferred technology used by U.S. coal-fired power plants for FGD purposes, primarily for bituminous coal. The wet FGDs can also achieve high removal efficiency of the oxidized Hg. Elemental mercury has very low solubility in wet FGD slurry and as a result, the elemental mercury leaves the wet FGD system along with the flue gas. Mercury speciation is a function of many parameters, but the main factors affecting the mercury speciation are halogen content of the flue gas and presence of selective catalytic reduction (SCR) systems for removal of nitrogen oxides (NO_x). As large numbers of EGUs have both SCRs and FGDs, the cost of mercury emissions compliance can be reduced significantly by utilizing the co-benefits of the existing AQCS components. As long as the elemental Hg concentration in the flue gas is below the MATS limit of 1.2 lb/TBtu or 4.0 lb/TBtu, the FGD systems can support MATS compliance efforts at very low operating cost {Yuan et al 4}.

Even with SCRs and FGDs, stack Hg emissions sometimes can be higher than the MATS emissions limit. This could happen due to factors affecting Hg speciation such as:

1) The oxidized Hg concentration in the flue gas is low due to SCR catalyst limitation. This can be due to parameters such as catalyst deactivation or plugging which severely affect the ability of SCR catalyst to oxidize mercury. This can be corrected by keeping the catalyst clean with equipment such as sootblowers or ash sweepers used in addition to sonic horns. The poisoning of the catalyst can be controlled by techniques such as catalyst formulation or combustion additives. Low mercury oxidation with or without SCR can be a result of low concentration of the halogen in the fuel. This is particularly observed on units firing PRB coal. This situation is however comparatively easy to correct by adding a small amount of halogen (such as iodine or bromine) to the flue gas to increase the overall mercury oxidation {Hinton et al 5}.

2) Another potential reason for stack Hg emission levels that are higher than the MATS limit can be due to the concentration of elemental Hg being higher at the stack as compared to the elemental Hg concentration at the FGD inlet. This scenario is more complicated and is not well understood. This phenomenon of higher elemental Hg concentration at the stack as compared to the FGD inlet is described as *mercury re-emission*. This phenomenon has been observed on units with wet FGDs only {Acharya et al 6}. This paper attempts to review our understanding of the re-emission process including: why re-emission takes place, what factors affect mercury re-emission, and what reliable techniques are available to control or eliminate mercury re-emission.

The occurrence of Hg⁰ re-emission was initially suggested by the data on chemical Redox processes occurring in the atmospheric cycle of Hg. It was determined that a potentially important step in the atmospheric cycle of Hg is the aqueous-phase (i.e., cloud and rain-water) reduction of Hg²⁺ by sulfite species [S(IV); the sum of HSO₃¹⁻ and SO₃²⁻ concentrations]. Kinetic data showed that the overall Hg²⁺ reduction rate was not sensitive to temperature changes from 60 to 85F and the reaction rate constant was inversely dependent on the S(IV) concentration in the range between 0.022 and 0.46 mM (millimolar) {Munthe, et al 7}. Conversely, kinetic data generated in another research effort indicated that the rate of Hg²⁺ reduction was proportional to S(IV) concentration and increased by more than one order-of-magnitude when the temperature was raised from 15 to 75F {Loon, et al 8}. Both sets of these contradictory kinetic data were

solely based on UV spectroscopic measurements of intermediate species, and not on quantitative Hg^0 emission data. Neither researcher considered the potential effect of hydrogen sulfide and its corresponding ions (S^{2-} and HS^-) in their kinetic experiments. Background atmospheric H_2S concentration, although small, may easily be comparable to that of mercury and could introduce sulfides into cloud chemistry and affect Hg^{2+} reduction reactions. The temperature of scrubbing liquor in wet FGD scrubbers is usually between 90 and 110F, which is quite different from the atmospheric conditions. The S(IV) concentrations in the scrubbing liquor can also be as high as several mM, which based on the contradictory atmospheric-science literature cited above, may inhibit or enhance the Hg^{2+} reduction process. Moreover, solids (mostly gypsum and to a lesser extent, fugitive fly ash) constitute a significant portion of the wet FGD slurry (up to 20%). The presence of fly ash may significantly affect Hg^0 re-emission. Solids were absent during all the atmospheric simulation studies.

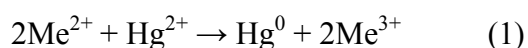
After the oxidized Hg is captured in the wet FGD solution, it can undergo three possible scenarios:

- 1) Hg can stay in the solution and leave the system along with the wet FGD blowdown.
- 2) The Hg is captured or precipitated along with gypsum or other solids and can exit the wet FGD along with gypsum.
- 3) The captured Hg is reduced to its elemental form, is reintroduced into the flue gas and leaves the wet FGD system.

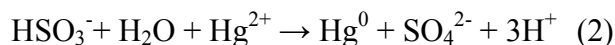
In a complex system (such as a wet FGD), all these scenarios can happen simultaneously to varying degrees. The unpredictability of the extent of Hg^0 re-emission in different wet scrubbers has prompted many fundamental research investigations {Chang et al 9}. These studies show that in the thick chemical soup that describes wet scrubber slurry, there exists a complex gas/liquid/solid-phase, electrolytic chemistry that controls the extent of Hg^0 re-emission and the partitioning of Hg between the gas, liquid, and solid phases.

B&W'S FUNDAMENTAL RESEARCH

B&W was the first organization to categorically identify Hg^0 re-emission from wet scrubbers {Farthing et al 10}. During work aimed at enhancing the mercury removal performance of wet FGDs, B&W researchers discovered that, under some circumstances, Hg^{2+} initially captured in a wet FGD system could be re-emitted in the form of Hg^0 . It was hypothesized that once Hg^{2+} dissolves and ionizes in solution, it is subjected to reaction with other dissolved constituents in the scrubber slurry. Principal among these may be metal ions such as the divalent ions of tin, manganese, iron, nickel, and cobalt, and soluble sulfite. Symbolically representing a metal ion as "Me," the following general reaction for the reduction of Hg^{2+} by dissolved metals was proposed:

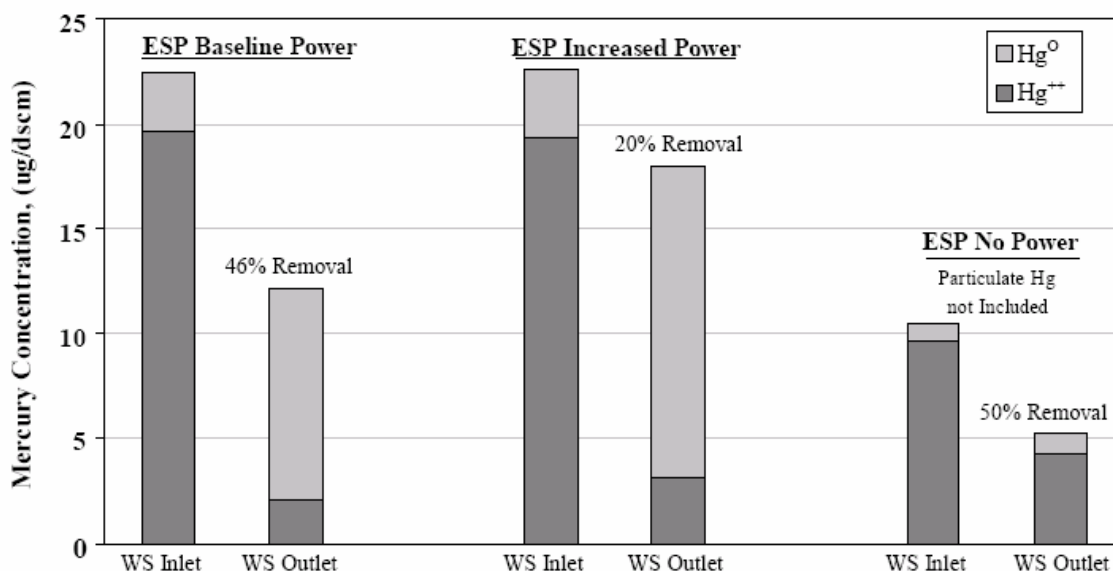


The reduction of Hg^{2+} by S(IV) (e.g., bisulfite) was proposed as follows:



The sources of metal ions in the scrubber are most likely the limestone, makeup water, and to a small extent, fly ash carryover to the scrubber from the dust collector. The soluble sulfite comes from the absorption of SO_2 . The objective of studies at B&W was then shifted to the development of techniques to counter the undesired Hg^0 re-emission from wet scrubbers. A series of follow-up experimental investigations were performed by evaluating Hg^0 re-emission with three different particulate matter (PM) control device/wet FGD configurations. Testing was performed to investigate the effect of electrostatic precipitator (ESP) operation on Hg re-emission. In the first configuration, a pilot-scale ESP was operated at its nominal condition. In the second configuration, the ESP voltage was increased by 60% above the baseline, and in the third operating condition, the ESP power was turned off. These tests showed that the operating voltage of the ESP had a direct, negative impact on the wet scrubber Hg control performance (see Figure 1) {Bailey et al 11}.

Figure 1. Effect of ESP Operating Voltage on B&W's Pilot-scale Wet FGD Hg Removal (Combustion of a Bituminous Coal, Ohio # 6) {Bailey et al 11}.

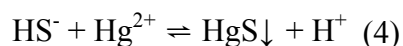
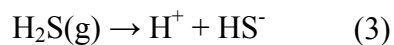


The proportion of gaseous Hg^{2+} and gaseous Hg^0 at the wet scrubber inlet was relatively the same for all three tests. However, for the high-power test, the amount of Hg^0 significantly increased across the wet scrubber. The outlet gaseous Hg^0 increased for the ESP baseline power test, but to a lesser extent than the high power case. No Hg^0 re-emission was observed when the ESP power was off. This indicated that the electric field somehow affected component(s) of the flue gas, which, in turn had a negative impact on Hg chemistry in the wet scrubber. The performance of the ESP is in turn dependent on coal composition, gas flux and also on ash properties. This co-relation of

ESP performance and mercury association with either solids or as dissolved in the scrubber slurry wet FGD was also highlighted in a previous paper {Brown et al 12}.

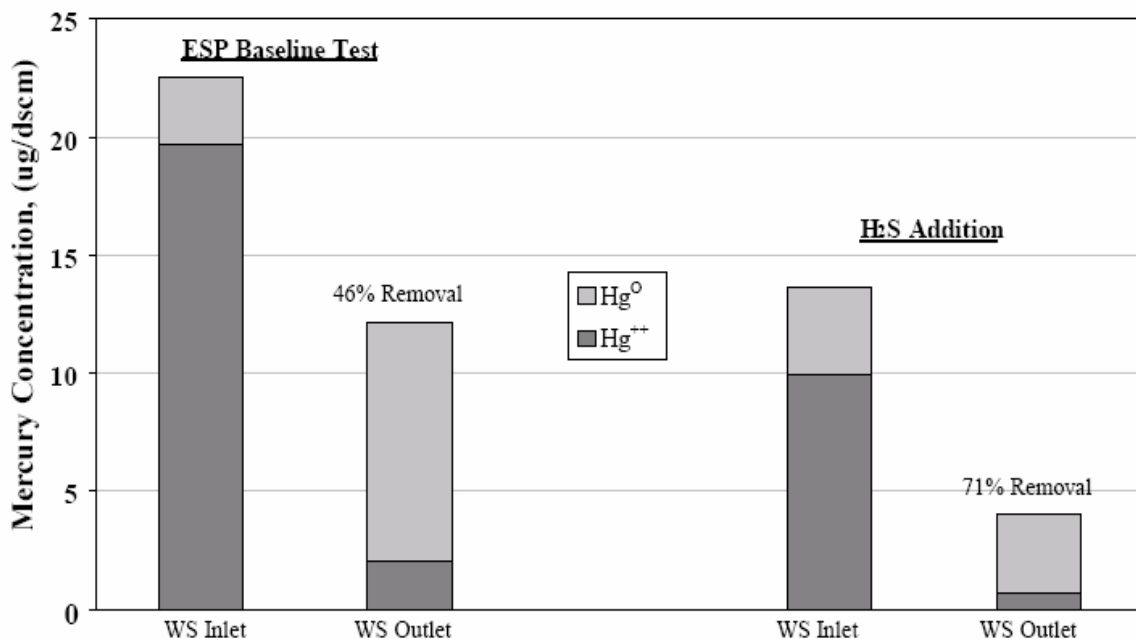
Further examination of the test data from B&W testing led to the following hypotheses:

- 1) Hydrogen sulfide (H₂S) or other reduced sulfur species could be present in coal combustion flue gases in trace amounts.
- 2) The amount of H₂S reaching the wet FGD system depends on both the conditions in the boiler and in the upstream particulate collector (high voltage generates ozone that can destroy H₂S).
- 3) H₂S reaching the wet FGD absorber can cause the precipitation of mercury as HgS at the gas-liquid interface by creating HS⁻ and S²⁻ ions. Precipitation of HgS effectively sequesters the mercury as an insoluble solid. The amount of Hg⁰ re-emitted from the wet FGD system was therefore seen as competition between the mercury reduction reactions previously described, and the precipitation of HgS described by the following reactions:



This hypothesis was experimentally tested during pilot-scale tests wherein H₂S was injected into the flue gases just upstream of a wet FGD system (see Figure 2). Hg⁰ re-emission present under baseline conditions was completely eliminated during the injection of H₂S.

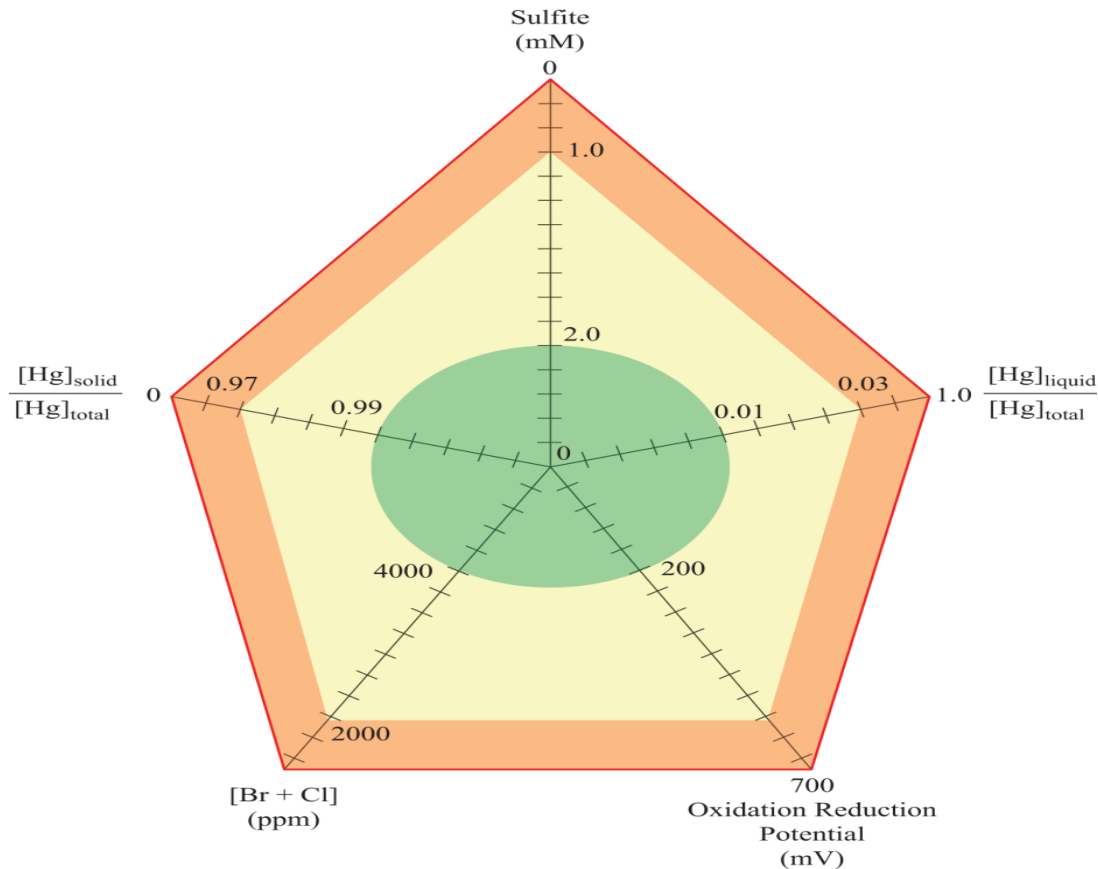
Figure 2. Effect of H₂S Injection on Wet FGD Mercury Control Performance, Pilot-scale Test {Bailey et al 11}.



B&W was subsequently awarded a patent for this approach (U.S. Patent 6,284,199). However, the injection of H₂S requires a system to generate the H₂S, a delivery system, and a distribution grid. To simplify the system and enhance safety, B&W devised an arrangement whereby a sulfide-donating liquid reagent (aqueous solution of sodium hydrosulfide, NaHS as one example) is injected into the suction side of the recirculation pump(s) of a wet FGD slurry in a manner specifically aimed at duplicating the impact of gas-phase H₂S. The injection of NaHS at this location causes a uniform distribution of the S⁻ and HS⁻ donating species in the spray zone of a wet scrubber. This method was tested in subsequent pilot-scale test campaigns and was shown to be as effective as the H₂S injection. From a practical point of view, this method is much preferable to the H₂S approach, and B&W was also awarded a patent for this approach as well (U.S. Patent 6,503,470).

In addition to B&W, other researches spent significant time to understand the mechanism of mercury re-emission from a wet FGD system. Many different parameters were investigated to understand their effects on Hg re-emission across the wet FGD, including: the halogen concentration in the scrubber slurry, the oxidation reduction potential (ORP) of the scrubber slurry, the form of occurrence of Hg in the slurry as Hg_{liq} or Hg_{solid}, and the effect of sulfite concentration. Figure 3 summarizes the effects of all these parameters on potential for Hg re-emission.

Figure 3. Effects of Operating Parameters on Mercury Re-emission {Berry S 13}.



Each axis in Figure 3 represents the parameter that can affect the mercury re-emission phenomenon. The green circle in the center represents operating conditions with low probability of Hg re-emission. These conditions include ORP around 200 mv, halogen concentration of around 4000 ppm, very low value of liquid phase Hg to total Hg ratio, and sulfite concentration of around 2.0 milimolar per liter (mM). All these parameters have been investigated in detail and their effects on Hg re-emission are well known. For example, it is well known that as the halogen concentration increases in the scrubber, the dissolved mercury can form complexes which are more resistant to being reduced as compared to the Hg-halogen complexes formed with low halogen concentration {Omine et al 14}.

The effect of ORP and re-emission is also well known. Usually higher ORP values indicate the presence of higher concentration of transition metals. Higher ORP also indicates that the Hg is in solution rather than associated with solids. As such, the high ORP increases the chances of mercury getting reduced due to reaction with transition metals and therefore getting re-emitted. In a paper from the Mega Symposium 2014, this co-relation of higher ORP or change in ORP with possibility of Hg re-emission was discussed {Brown et al 12}. This paper showed that when mercury was associated with solids, then the probability of re-emission was lower as compared to the case when mercury was in the solution. The role of sulfites in suppressing Hg re-emission has also been investigated extensively and as indicated in Figure 3, higher sulfite concentration helps to reduce the potential of mercury re-emission. All of these parameters indicate that Hg re-emission can be mitigated by controlling the scrubber operating parameters. The problems with controlling scrubber operating parameters to control mercury re-emission is that it can lead to some other issues such as sulfite blinding or an increase in the rate of corrosion when using the halogen concentration as a parameter. As a result, the most commonly used method for mitigating mercury re-emission from wet FGDs is the addition of a sulfide-donating liquid reagent, such as an aqueous solution of sodium hydrosulfide (NaHS), as one example.

SODIUM HYDROSULFIDE (NaHS) FIELD TESTING

B&W's patented concept of injecting sulfide-donating liquid reagent (such as NaHS) has been demonstrated successfully to control mercury re-emission. As a result, NaHS injection into a wet FGD system can be used to support MATS compliance efforts. Testing at many different units, both in the U.S. and internationally, during the last ten years has demonstrated the capability of NaHS to mitigate Hg re-emission irrespective of the type of wet FGD system. As the understanding of NaHS injection and its effects on Hg re-emission control progressed, the injection rate of the additive has been reduced significantly. Data presented at the 2016 Electric Utility Environmental Conference (EUEC) showed that co-injecting calcium bromide along with NaHS at very low injection rates was able achieve targeted Hg emissions levels {Flowers et al 15}. Testing was performed at Texas Municipal Power Agency's (TMPA) 450 MW Gibbons Creek Unit located in Anderson, Texas. The PRB coal-fired unit has ESP and wet FGD as AQCS equipment. The objective was to achieve MATS compliance using a halogen addition to the coal and a sulfide-based chemical for preventing Hg re-emission. The halogen used for achieving mercury oxidation was calcium bromide. Three commercially available sulfide-based mercury re-emission controls were compared to quantify their effectiveness and cost.

Figure 4 shows that the stack total Hg (shown by the dark red line) under baseline operation was around 4 to 6 lb/TBtu with average wet FGD inlet Hg at 12 to 15 lb/TBtu (shown as light blue dots). Halogen injection started at around 50 ppm of bromine added to the coal and eventually was increased to 100 ppm of bromine (shown by the purple line). The initial NaHS injection rate was around 14 g/hr and was later reduced to a steady operation rate of about 6 g/hr over the test period (shown by the black line). It was observed that a large NaHS injection rate was initially required to de-saturate the scrubber solution. Once the driving force for removal of oxidized Hg as HgS is created, then a lower maintenance dose of NaHS can be used to make sure that the scrubber solution stays de-saturated with respect to oxidized mercury. It can be observed that during the test period from 7/12/15 through 7/24/15 with injection of CaBr₂ and NaHS, the unit was always in MATS compliance. The plant also tested two other sulfide-based wet FGD additives. The comparison of the three additives is shown in Table 1.

Figure 4. TMPA Gibbons Creek NaHS Injection Testing Data {Flowers et al 15}.

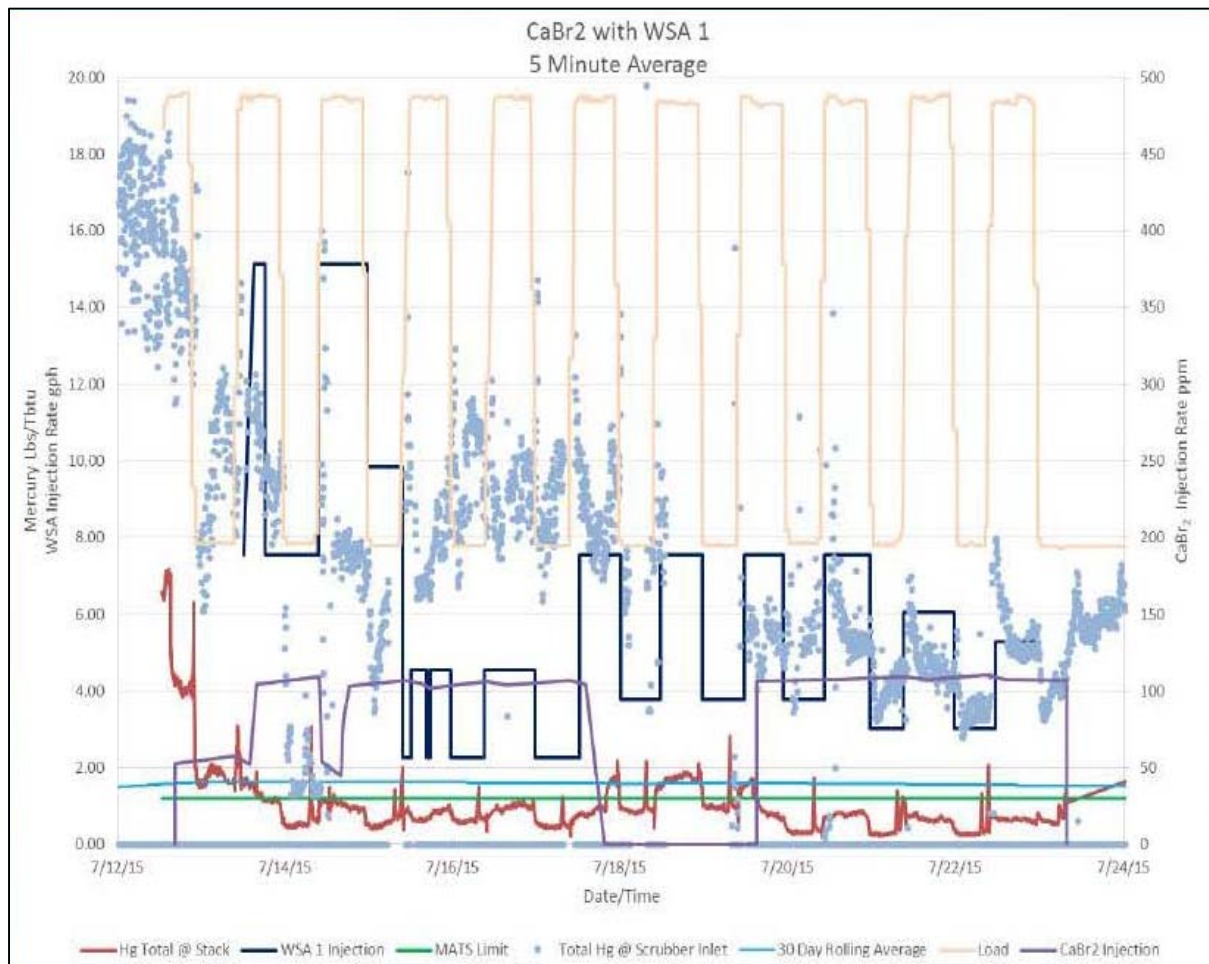


Table1. Compliance Cost Comparison {Flowers et al 15}.

Chemical	Quantity in Gallons	Total Cost \$
WSA1 or NaHS	1,183	13,131
CaBr ₂	2207 (with high wet FGD inlet Hg of average of 9 lb/TBtu during the test period)	22,071
Total		35,202
WSA2 or Hydro polysulfide	2,200	40,937
CaBr ₂	1056 (with high wet FGD inlet Hg of average of 7.0 lb/TBtu during the test period)	10,566
Total		51,503
WSA3 or Ferrous Sulfide	3,703	32,995
CaBr ₂	759 (with high wet FGD inlet Hg of average of 6.0 lb/TBtu during the test period)	7,595
Total		40,590

As seen from Table 1, the overall cost for the NaHS + CaBr₂ combination was the lowest amongst all three cases. The higher CaBr₂ cost during the NaHS period was due to the fact that the wet FGD inlet Hg concentration was much higher during the NaHS trial. NaHS or any other sulfide does not increase or decrease the halogen injection quantity and the rate of injection is mainly based on coal Hg and other flue gas factors. As such, the plant decided to use CaBr₂ + NaHS as their MATS Hg emissions compliance strategy. As demonstrated here and at other trials, the sulfide-based additives can be very effective in helping to achieve targeted Hg limits provided that the wet FGD inlet elemental Hg concentration is lower than the MATS limit of 1.2 lb/TBtu. Therefore, many plants firing bituminous and PRB coals, with or without an SCR, are using CaBr₂ as the halogen of choice for achieving high mercury oxidation.

With the use of CaBr₂ for mercury oxidation, some balance-of-plant effects have also been observed. The most commonly observed effect has been air heater corrosion, primarily of the cold end basket. B&W, along with Chem-Mod LLC, performed a comparison of iodine and bromine to evaluate their Hg oxidation capability, corrosion potential and cost. The testing was performed at Unit 1 of the Whelan Energy Center in Hastings, Nebraska. This unit has a capacity of 80 MW and is equipped with ESP for particulate control. The mercury oxidation was compared using EPA method 30B sampling at the air heater outlet. The corrosion potential was compared using electrochemical noise (ECN) probe technique which has the capability to measure the rate of corrosion while the unit is online. Table 2 shows the results from that comparison testing. All of the halogen injection tests were performed for a 12-hour period to obtain reliable data. Metallurgy of the air heater basket was simulated using the ECN probe.

Table 2. Halogen Performance Comparison {Gadgil et al 16}.

Test	% Hg Oxidation	Rate of Corrosion Mils/year	Testing Cost in \$ for 12 hours of Halogen Injection
Baseline	40	0.16	N/A
75 ppm Br or 0.75 g/hr	83	0.39	90
150 ppm Br or 1.5 g/hr	94.5	1.2	144
10 ppm iodine 0.1 g/hr	93.1	0.16	72
25 ppm iodine 0.25 g/hr	98.5	0.31	180

As seen from Table 2, both bromine and iodine were very effective in terms of Hg oxidation capability. To achieve high Hg oxidation, 150 ppm of bromine was needed, whereas the same percentage of mercury oxidation was obtained with only 10 ppm of iodine. The most important aspect of this study was the corrosion rate measurement. With 10 ppm of iodine, the corrosion rate was the same as the baseline corrosion rate but with 93% Hg oxidation. With bromine at 150 ppm injection, the oxidation rate was 94%, but the rate of corrosion was almost 10 times higher as compared to the baseline rate. This data indicates that iodine could be a better alternative to bromine for Hg oxidation.

The recent U.S. EPA effluent limitation guidelines (ELG) now regulate the wastewater discharge from power plants. It has been demonstrated that halogens such as bromine, iodine, and chlorine can lead to formation of tri halogenated methane (THM) in water, a known carcinogen. THM formation is related to the halogen application rate, and as the rate of halogen injection was lowered, the THM concentration was reduced proportionately {Abbott et al 17}. Even on units with SCRs, the rate of halogen injection is very sensitive to the overall catalyst activity and can change significantly over the catalyst operating period as the catalyst deactivates. Therefore, in addition to requiring lower reagent consumption rates and experiencing lower air heater corrosion rates, lower THM formation is another reason that iodine is a better choice than bromine to achieve Hg oxidation.

CONCLUSIONS

The Mercury and Air Toxic Emissions Standards (MATS) rule regulate that all EGUs in the U.S. must limit stack Hg emission to below 1.2 lb/TBtu or 4.0 lb/TBtu, depending on fuel. Many power plants are using existing AQCS equipment as the lowest operating cost MATS compliance strategy. As such, reliable oxidation of Hg and removal of oxidized Hg by FGD has become very important. On units equipped with wet FGDs, the phenomenon of Hg re-emission can increase Hg emissions. Factors such as ORP of the scrubber slurry, halogen concentration in the scrubber slurry, the ratio of dissolved Hg to total Hg in scrubber slurry, and the sulfide concentration in the slurry affect the Hg re-emission process. Out of these parameters, the sulfide concentration is the easiest and least costly parameter that can be controlled as a strategy to eliminate Hg re-emission. During pilot-scale tests aimed at enhancing the mercury removal performance of wet FGD systems, B&W discovered the important role of sulfides on the sequestration of soluble ionic mercury (Hg^{2+}) and prevention of elemental mercury (Hg^0) re-emission. B&W was subsequently awarded

patents utilizing the sulfide chemistry and process for suppression of Hg reemission in wet FGDs. Implementation of the B&W patented technology that utilizes a sulfide-donating liquid reagent such as sodium hydrosulfide (NaHS) on several operating wet scrubbers has been very successful. Recent testing of different sulfides including NaHS demonstrated the advantages of NaHS compared to other sulfides available in the market for Hg re-emission control. The use of halogens to maintain high Hg oxidation is very important even on units equipped with SCR systems. Though bromine has been the commonly used halogen for Hg oxidation, recent test data indicates that use of iodine for mercury oxidation can offer many advantages over bromine. Based on the analysis of the data collected over the last twenty years, the use of sodium iodide and sodium hydrosulfide can be a cost-effective and reliable combination for Hg emissions reduction within an overall MATS compliance strategy.

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