

Estimates of Increased Black Carbon Emissions from Electrostatic Precipitators during Powdered Activated Carbon Injection for Mercury Emissions Control

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1 Abstract:

The behavior of mercury sorbents within electrostatic precipitators is not well-understood, despite a decade or more of full-scale testing. Particulate filters have shown evidence that powdered activated carbon (PAC) can penetrate ESPs significantly during sorbent injection for mercury emissions control. Recent laboratory results suggest PAC collection behaviour in ESPs that is different than fly ash. The present analysis considers a range of assumed differential ESP collection efficiencies for PAC as compared to fly ash. Estimated emission rates of PAC are compared to estimated emission rates of black carbon on fly ash, each corresponding to its respective collection efficiency. To the extent that any emitted PAC exhibits size and optical characteristics similar to black carbon, such emissions could increase black carbon emissions from coal-based stationary power generation. The results reveal that even for the low injection rates associated with chemically impregnated carbons, black carbon emissions can easily double if the fine fraction of the native fly ash has a low LOI. Increasing sorbent injection rates, larger collection efficiency differentials as compared to fly ash, and decreasing sorbent particle size all lead to increases in the estimated black carbon emissions.

2 Introduction

Among the various strategies for reducing mercury emissions from coal combustion, injection of powdered sorbents has been extensively tested and demonstrated at full-scale. Although most such tests have been conducted in the U.S., international momentum is growing as well. In 2009, the Governing Council of the United Nations Environment Programme (UNEP) approved Decision 25/5, mandating the pursuit of a global, legally binding instrument for reducing mercury emissions into the environment. A large majority of sorbent injection tests have involved injection of mercury sorbents upstream of an electrostatic precipitator (ESP). This reflects the dominance of ESPs in use as particulate control devices at coal-fired power plants: Approximately 70% of plants in the U.S. (1), 95% of plants in India (2), 55% of plants in Russia (3) and 88-90% of plants in China (4) use ESPs for particulate matter (PM) control. Despite the predominance of ESPs installed at coal-burning power stations, the challenging experimental environment they

present has prevented detailed, systematic examination of how mercury sorbents behave.

ESP collection efficiency is known to be highly dependent on the resistivity of the particulate matter (PM). Whereas optimum values of resistivity range from 10^8 - 10^{12} ohm-cm and values for FA from coal combustion typically fall in or near this range, PAC has a value of ~ 1 ohm-cm (5), indicating lower ESP collection efficiencies than those for FA, though the degree of difference has not been determined. Full-scale sorbent injection test results are mixed on the issue. Full-scale PAC injection testing at the Brayton (6), Meramec (7), Monroe (8) and Pleasant Prairie (9) sites did not negatively impact stack opacity. However, testing of 18 different sorbents at Conesville (10) resulted in increased ESP sparking, decreased ESP power, or increased opacity in most cases. At Stanton Unit 1 (11), particulate filters used in conjunction with gas sampling at the ESP outlet showed dramatic differences in coloration, though these may have reflected load changes. At the Lausche site (12), observed opacity increases were highly

dependent on particle size and injection rate. Injection of PAC with a mass median diameter (MMD) of 20 μm yielded a constant opacity of 5% for injection rates up to 8 lb/MMacf. However, opacity nearly doubled to 9% when MMD was reduced to 5 μm (at 2.5 lb/MMacf), and more than tripled to 15-16% when MMD was reduced to 1 μm (at 1.5 lb/MMacf).

The penetration of injected PAC through ESPs is problematic to the extent that 1) PAC emitted into the atmosphere behaves like combustion-derived black carbon (BC), 2) compared to BC emissions into the atmosphere due to unburned carbon on fly ash, significant increases occur during PAC injection, and 3) the climate forcing potential (CFP) of emitted BC (both PAC and on FA) is either significantly positive or significantly negative compared to the CFPs of other constituents emitted from the same source. In this regard, there is an obvious convergence: coal combustion represents the potential for large emissions of both mercury and CO_2 , and in the case of PAC injection, the potential exists that efforts to reduce emissions of the former will offset reductions of the latter. The present analysis seeks to estimate the relative importance of PAC collection efficiency within and PAC penetration through an ESP and the resultant impacts on emitted BC from coal combustion.

3 Methodology

The analysis uses the 2007 U.S. electric power generation from coal (2016.1×10^9 kWh (13)) to infer total flue gas volumetric flow rates, based on an assumed flue gas production rate of 323 m^3/GJ (14). Though the present analysis calculates total flue gas production from total annual power generation (2016.1×10^9 kWh (13)), this differs only by a factor of two from similar calculations based on total annual coal consumption (1.145×10^9 short tons (13)) assuming a Utah #10 bituminous coal (6). Applying PAC injection at representative injection rates (lbs/MMacf) then gives the total mass of PAC injected nationwide. Basing the analysis on an assumption of PAC injection applied to the total U.S. electric power generation from coal admittedly trades precision for results of a more general nature. However, this is justified based on the large variability in CFPP configurations and operating conditions, as well as the high degree of uncertainty in the BC content of emitted FA (see additional discussion in Results). It is acknowledged that PAC injection is but one of several options available

to unit operators intending to implement mercury emissions reductions.

ESP performance is considered in terms of an overall PM collection efficiency (both FA and PAC), a collection efficiency differential between the coarse and fine fractions of the PM (both FA and PAC), and finally a collection efficiency differential between the fine fraction ($< 1\mu\text{m}$) of the FA and the fine fraction of the PAC. While reasonable values for the overall ESP collection efficiency exist, the separate values for the collection efficiency differential between coarse and fine fractions of the PM, and between the fine FA and the fine PAC, are unknown. It was determined that the collection efficiency differential between the coarse and fine fractions of the PM (both FA and PAC) affected the absolute PM and BC emissions, but did not affect the percentage increase in BC emissions. The analysis assumes a 32% collection efficiency differential between the coarse and fine fractions of the PM (i.e., fine fraction collection efficiency of 67.5%) (6). The analysis assumes only negative values for the collection efficiency differential between the fine FA and the fine PAC (i.e., ESP performance reductions for PAC), based on the knowledge that PM resistivity is a key factor in ESP performance, and that the value for PAC (~ 1 ohm-cm) (5) falls well outside of the optimum range ($10^8 - 10^{13}$ ohm-cm) (15).

To estimate BC emissions with FA emitted from ESPs, the analysis assumes a 2007 U.S. coal consumption of 1.145×10^9 short tons, having 10% ash content, a 40% bottom ash, 60% fly ash split, and 0.5% of the fly ash in the fine fraction ($< 1\mu\text{m}$) (6). The ESP removes the coarse fraction of the FA from the flue gas according to the overall ESP collection efficiency (99.5%). Although it is assumed that injected PAC will agglomerate with FA particles, and that the degree of climate forcing exerted by BC depends on to what degree it is externally or internally mixed within an agglomerate (16), the present analysis only considers the increase in total BC escaping an ESP due to PAC injection. Although wet scrubbers may be present downstream of an ESP, the analysis assumes that the particle capture dynamics in a wet scrubber are sufficiently similar between FA and PAC that the same collection efficiency applies for both, thus yielding the same percent increase in BC emissions with or without a wet scrubber, although absolute BC emissions would be lower for an ESP followed by a wet scrubber than for an ESP alone.

4 Results

The percentage increase in BC emissions chiefly depends on the PAC injection rate, which is controlled, and the ESP collection efficiency differential between FA and PAC, which is both unknown in a fundamental sense and likely to be highly variable and site-specific. The estimates of BC emissions due to PAC injection were determined to be independent of the ESP overall collection efficiency or the ESP collection efficiency differential between the coarse and fine fractions of the PM, where PM is a mixture of FA and PAC. Figure 4-1 shows estimated changes (increases) in BC emissions from ESPs, subject to the assumptions stated above, as a function of both PAC injection rate and the differential ESP collection efficiency between PAC and FA. Both panels in Figure 4-1 assume injection of conventional PAC, i.e., PAC that is neither finely ground nor chemically impregnated, and assume representative injection rates accordingly. Because of advances in mercury sorbent development, the results for conventional PAC in Figure 4-1 are primarily of historical interest; BC emissions estimates for more advanced

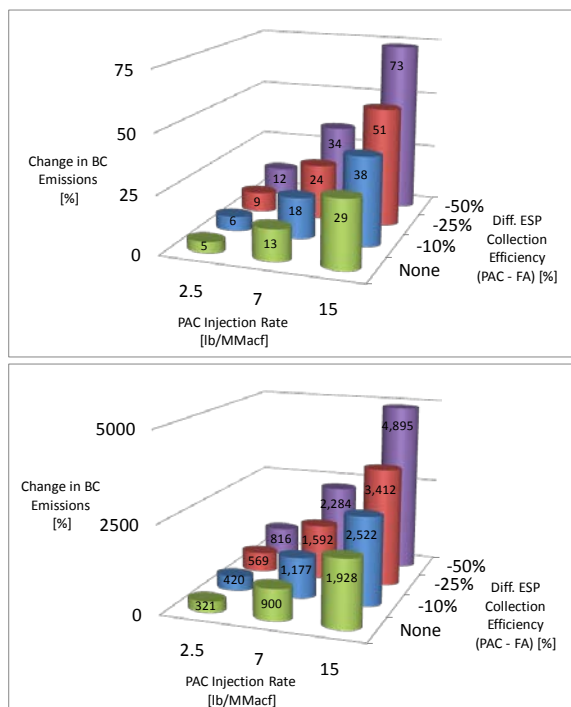


Figure 4-1: Estimated increases in BC emissions from ESPs during sorbent injection using conventional PAC. Upper: Assumed 40% BC content in submicron FA fraction (18). Lower: Assumed 0.6% BC content in submicron FA fraction (19) and references therein).

PAC formulations are shown in Figures 4-2 and 4-3. The BC content of the fine fraction ($< 1 \mu\text{m}$) of the FA is not well known, and thus the upper and lower panels in Figure 4-1 represent two assumed values. The top panel of Figure 4-1 assumes a 40% BC content in the fine FA fraction (18). The associated estimates of BC emission increases during injection of conventional PAC (Figure 4-1) vary from 5 to 29% for the most optimistic scenario of no differential in collection efficiency between FA and PAC within an ESP. The worst case scenario, i.e., the highest sorbent injection rate and highest collection efficiency differential, yields an estimated 73% increase in BC emissions. By comparison, the bottom panel of Figure 4-1 presents estimates of BC emissions for conventional PAC injection assuming only 0.6% BC content in the fine FA fraction (19) and references therein). Clearly, the assumption of much lower BC content in the FA leads to PAC injection having a much greater impact on estimated BC emissions: The most optimistic scenario (no collection efficiency differential, lowest PAC injection rate) produces an estimated 321% increase in BC emissions, i.e., roughly a quadrupling of BC emissions.

Newer mercury sorbent formulations, particularly bromine-impregnated PACs, have repeatedly shown much improved performance over conventional PACs, achieving mercury removal efficiency targets under the most challenging conditions (e.g., high concentrations of SO_3) and at much lower injection rates. Figure 4-2 shows estimates of BC emissions increases for brominated PAC injection. The underlying assumptions applied for brominated PAC injection were, 1) identical particle size distribution as conventional PACs, 2) lower injection rates than conventional

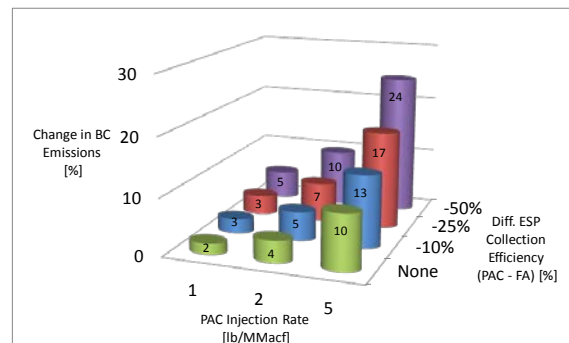


Figure 4-2: Estimated increases in BC emissions from ESPs during brominated PAC injection. Assumed 40% BC content in submicron fraction of fly ash (18).

PACs, and 3) no impact of bromination on ESP collection efficiency differential (based on lab-scale results (20)). Estimated increases in BC emissions for brominated PAC injection under the most optimistic scenario (no collection efficiency differential, 40% BC content in fine FA fraction) vary from 2 to 10%, with a maximum 24% increase in BC emissions at the highest injection rate (5 lb/MMacf) and for the highest collection efficiency differential (-50%). As noted earlier, assuming a lower BC content for the fine FA fraction greatly increases the impact of PAC injection on BC emissions. For a BC content of 0.6% in the fine FA fraction, the most optimistic scenario (no collection efficiency differential) for brominated PAC injection yields BC emissions increasing from 129 to 643%, i.e. a two to seven times increase in BC emissions.

In addition to chemical impregnation, more finely ground powdered sorbents have been developed, or produced on-site, in order to increase available external sorbent surface area to reduce mass transfer resistance. However, finely ground PACs have generally not shown any improved performance over more coarse products of the same chemical formulation (12). One potential explanation that has been put forth has been that powdered sorbents tend to agglomerate to various degrees during the feeding and injection process, depending on the sorbent and the length and materials of construction of the feed line. Agglomeration can be promoted by either by high particle mass loading or by triboelectric charging during pneumatic feeding, effectively shifting the particle size distribution (PSD) to a more coarse state. The effects of such phenomena on the sorbent PSD during pneumatic feeding have been demonstrated at lab scale (21) and has subsequently been observed during full-scale testing (22) (23). Although these effects have so far limited the performance of finely ground sorbents, it is possible that sorbent formulations or sorbent feeding methods could progress such that such fine powders could be successfully injected into the flue gas, thereby providing mass transfer advantages for mercury capture. Figure 4-3 presents estimates of BC emissions increases associated with the injection of a finely ground brominated PAC, assuming a 40% BC content in the fine FA, in alignment with the estimates shown in Figure 4-1 (upper panel) and Figure 4-2. Particle size analysis (Particle Technology Laboratories, Downers Grove, IL) indicates that a conventional PAC such as Norit DARCO FGD with a mean particle size of 18-25 μm has approximately 3.5% of its mass

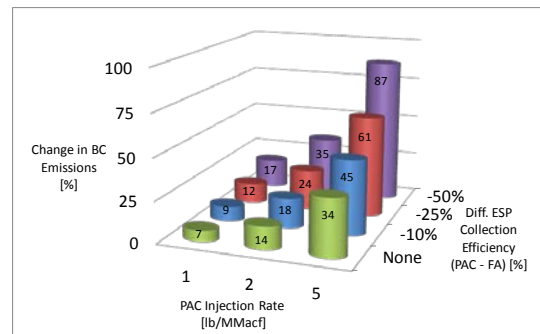


Figure 4-3: Estimated increases in BC emissions from ESPs during the injection of finely ground, brominated PAC. Assumed 40% BC content in submicron fraction of fly ash (6).

in particles less than 1 μm in size (24). By comparison, the measured PSD for a finely ground PAC with a mean particle size of 6 μm shows 12.5% of the mass in particles less than 1 μm in size (23). Figure 4-3 shows that for finely ground brominated PAC under the most optimistic scenario of no collection efficiency differential and 40% BC content in the fine FA fraction, BC emissions increased from 7 to 34% as compared to the 2 to 10% increases estimated for the more coarse brominated PAC in Figure 4-2. In the worst-case scenario of 5 lb/MMacf injection rate and -50% ESP collection efficiency differential, BC emissions increased by 87%. Assuming a lower BC content for the fine FA fraction (0.6%) yields estimates of BC emissions increases that are generally two orders of magnitude larger than those shown in Figure 4-3.

It is important to place these estimated BC emissions increases into context. The radiative forcing of all BC, including direct forcing, snow/ice albedo decrease, and indirect forcing, is complex and less well understood than the climate effects of CO_2 . Though the 2007 Assessment Report of the Intergovernmental Panel on Climate Change (IPCC) (17) estimates the direct forcing effects of CO_2 , BC, and CH_4 to be 1.66, 0.05-0.55, and 0.48 W/m^2 , respectively, Ramanathan and Carmichael (25), and references therein arrive at a BC direct forcing value of 0.9 W/m^2 (ranging between 0.4 to 1.2 W/m^2), which they assert would be second only to CO_2 in magnitude and greater than all other GHGs. The percentage of all BC emissions globally that are attributable to fly ash emitted from coal-fired power plants (CFPPs) ranges from 10% (26) to less than one-tenth of one percent (19), which represents still more uncertainty in the baseline BC emissions on which the BC emissions increases calculated herein are based. The global estimates of BC emissions are dominated by intentional and accidental

burning of forests and savannahs, which together account for about 37% of all global BC emissions (19) (26). On-road diesel-fuelled transportation (approximately 13%) and residential wood combustion (approximately 11%) are also large sources (19) (26). However, emissions from these various combustion sources can consist of organic carbon (OC), which tends to have a cooling effect on climate (negative climate forcing), or elemental carbon (EC), which can have warming effects (positive climate forcing). Although biomass burning dominates total BC emissions globally, BC emissions from such sources tend to be primarily OC, whereas BC emissions from internal combustion sources tend to be EC.

On a global scale, BC emissions associated with PAC injection would appear to constitute a potentially large increase in a fairly small source category for a constituent whose climate forcing potential is significantly smaller than that of CO₂. However, several additional factors warrant consideration. First, BC emissions from PAC injection are easier to control than any other BC source category because the technology is not yet in widespread use, even in the U.S., and therefore optimization with respect to BC emissions is still possible. Second, unlike many BC source categories, PAC injection is an ancillary process, not essential for livelihood or sustenance, greatly diminishing the type of ethical dilemma that can arise when considering reduction of BC emissions from other sources. Third, compared to global estimates, the percentage of U.S. BC emissions attributable to CFPPs falls toward the higher end of the range: 8% of all BC emissions in the U.S. are estimated to originate from CFPPs (27). Further, in the 15 year interval between 1990 and 2005, BC emissions in the U.S. decreased by 30%, and are projected to decrease by a further 80% (from 2005 levels) by 2030 due primarily to the adaptation of PM controls for transportation diesel engines (27). This alone would double the contribution that CFPPs make to total BC emissions in the U.S. to 16% before considering any potential BC emissions increases from PAC injection. Cast against such strong trends of decreasing BC emissions in the U.S., the potential for mercury emissions controls to lead to sharp increases in BC emissions would not likely go without notice.

Outside the U.S., the movement of the United Nations Environment Programme (UNEP) toward a binding global treaty on mercury in the environment increases the likelihood that other countries will join the U.S. in placing

limits on mercury emissions from CFPPs. India and China each have very large coal-based power generating capacity, with ESPs installed on the large majority (88-90% in China (4) and 95% in India (2)). To the extent that the ESP collection efficiency differential between FA and PAC increases with decreasing general ESP performance, implementing PAC injection upstream of a poorly maintained ESP, even at low injection rates, would result in the greatest increases in BC emissions. Whereas the present analysis assumed an overall ESP collection efficiency of 99.5%, older ESPs in the U.S. have collection efficiencies as low as 90% (28); similarly, two-thirds of ESPs in the countries comprising the Newly Independent States of the former Soviet Union (NIS) exhibit collection efficiencies as low as 88% (3). Sorbent injection technology is typically promoted for its low capital and operating costs. Such cost estimates would likely increase if curtailing BC emissions during PAC injection required ESP upgrades, sorbent switching or formulation changes, or downstream installation of polishing wet scrubbers or fabric filters. Non-carbon sorbents, which tend to be light gray in color, have shown precipitation behavior in lab-scale tests that preliminarily appears more similar to FA than PAC (20), but their behavior at full-scale remains unknown. As noted earlier, while a downstream WFGD would capture additional PM (both FA and PAC), the removal efficiency will likely be the same for both the fine FA and fine PAC, lowering absolute PM emissions but leaving the percentage increases in BC emissions unchanged.

Another potential concern, though more speculative in nature, is the concentration of mercury and other condensable metals adsorbed onto the fine fraction of the PAC escaping the ESP. Recent lab-scale electrostatic precipitation studies (20) suggest that during electrostatic precipitation of PAC and FA mixtures, PAC preferentially collects on the discharge electrodes. Because less PM collects on discharge electrodes they are typically programmed for longer rapping intervals, leading to longer exposure of the collected PM to the flue gas. To the extent that rapping of an upstream discharge electrode leads to resuspension of the collected PM and preferential collection of its PAC fraction on downstream discharge electrodes, one would expect the PM at the outlet of an ESP to be significantly enriched in PAC. The combination of longer exposure to the flue gas and greater adsorption capacity could lead to much higher concentrations of condensable metals on any fine PAC escaping the ESP. Thus, in addition

to assessing the basic precipitation behavior of PAC (and other mercury sorbents), and evaluating the optical properties of non-carbon sorbents, it is also recommended that residual mercury sorbent material found penetrating an ESP should subsequently be analyzed to determine the concentrations of condensed species that they contain.

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