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Reduction of mercury emissions from coal fired power plants

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Introduction
Coal fired power plants (CFPP) are the largest anthropogenic stationary sources of mercury emissions in Europe. The USA has developed policies to reduce emissions of mercury from CFPP. The current HM Protocol also addresses the emissions of mercury from CFPP. The Protocol provides information on the Best Available Techniques (BAT) to reduce emissions of mercury. The Protocol does not give a specific emission limit value (ELV) for mercury emissions from CFPP. With the revision of the current protocol new standards aimed at reducing mercury emissions from CFPP could be amended to the HM Protocol now in revision.

This paper discusses current mercury emissions from stationary coal fired power plants, possible emissions abatement techniques, costs of mercury abatement, current policies and legislation in the UN ECE region and the proposal for an emission limit value to be added to the HM Protocol.

Emissions of mercury from coal fired power plants

The global mercury emission in 2008 was about 8000 tons, 36% of which (2900 tons) is originating from anthropogenic activities. The main anthropogenic source for mercury emissions is the combustion of fossil fuels. Table 1 gives an overview of the mercury emission per continent from stationary combustion sources in 2008, as presented by Pirrone at the CEM conference in 2009.

At the 47th meeting of the WGSR in September 2010 TNO and MSC East will present the results of new estimates of emissions of heavy metals in the UN ECE region for different scenarios. TNO estimated the emissions of mercury from power plants to be 119 ton/year, for UNECE /Europe in 2010. This is 47% of the total mercury emissions in UNECE- Europe.

Table 1: Global emissions of mercury in 2008 from stationary combustion plants (ref. 1)

Continent	tons Hg
Europe	89
Africa	205
Asia + Russia	905
North America	80
South America	31
Australia and Oceania	113
Total	1423

Emissions of mercury from CFPP for the EU are also reported to the E-PRTR database. Total reported mercury emissions from stationary sources in 2008 are 37.8 t/a, of which 21.2 tonnes are from CFPP (ref. 14). This is about a quarter of the emissions estimated by Pirrone. It is unclear whether all emissions are reported to E PRTR. CFPP are however the largest stationary source of mercury emissions in E PRTR. The reported emissions per installation can be found at the E-PRTR website, (ref. 14).

Because of the uncertainty in the mercury emissions an estimate was made on the basis of reported coal consumption for power production. Table 2 gives an overview of the coal consumption burned for the generation of electric power in 2008 (ref. 26). The mercury emission is estimated based on the usual range of mercury content of coal (ref. 4). The mercury emission reduction caused by abatement techniques is not taken into account, so the third column displays the worst case scenario. As a comparison the table also presents the reported mercury emission and the number of power plants of Europe according to the e-PRTR-database 2008.

	USA Energy Information administration			e-PRTR 2008	
Region	Coal consumption Mton coal/a	Calculated Emission ton Hg (0.05 g Hg/ ton coal)	Calculated Emission ton Hg (0.2 g Hg/ ton coal)	Reported emission ton Hg	number power plants
Europe	1032	52	206	21.2	193
Eurasia	457	23	91	0.50	2
Total Europe + Eurasia	1489	74	298	21.7	197

Table 2. Estimated mercury emissions based on coal consumption

The emission estimated by TNO of 119 ton/y is well within the range of estimated emissions based on coal consumption.

The USA Department of Energy estimated emissions of mercury from CFPP in the USA to be 48 tonnes in 2005 (ref. 15).

Abatement of mercury emissions from coal fired power plants

The mercury content of coal is typically in the range of 0.05-0.2 g/ton (ref. 2). Mercury is volatilized during combustion and converted to gaseous elemental mercury Hg(0). Subsequent cooling of the flue gas and interaction of Hg(0) with other flue gas constituents, such as chlorine and unburned carbon, results in partial oxidation of the Hg(0) to gaseous oxidized forms of mercury Hg(2+) and particulate-bound mercury Hg(P). As a result, coal combustion flue gas contains varying percentages of Hg(P), Hg(2+), and Hg(0). The exact speciation of mercury has a profound effect on the Hg capture efficiency of existing air pollution control device (APCD) configurations, which has been found to range from 0 to over 90 percent.

For a coal fired power plant APCD normally consist of several abatement techniques. In most cases an ESP is used as a first step in reduction of dust emissions. More and more installations also apply a fabric filter to further reduce emissions of dust. Most installations in the EU and part of the installations in the USA and Canada reduce emissions of SO_2 by applying flue gas desulphurization (FGD) based on wet or semi-dry scrubbers. In many modern power plants also selective catalytic reduction (SCR) is used to reduce emissions of NO_x.

The Hg(P) fraction is typically removed by a particulate control device such as an electrostatic precipitator (ESP) or fabric filter (FF). The Hg(2+) portion is water-soluble and therefore a relatively high percentage can be captured by the wet flue gas desulphurization (FGD) systems. The Hg(0) fraction is generally not captured by existing APCD. However, when an SCR is applied this will promote oxidation of Hg(0) to Hg (2+)and enhance Hg capture across a downstream FGD (ref. 3).

<u>Mercury abatement as co-benefit of reduction of NO_x, SO_2 and dust</u>

The EU-BREF for Large Combustion Plants presents techniques to reduce mercury emissions from coal fired power plants. These are based on a combination of techniques that are designed to reduce other pollutants, i.e. to reduce SO_2, NO_x and particulate matter (PM). This approach is in line with the above mentioned mechanisms. The BREF Large Combustion Plant gives the following conclusions on the use of BAT (ref. 25).

- A fabric filter or ESP removes about 50% of the mercury from the flue gas.
- A FGD removes 50% of the residual mercury emission. This means that when an ESP and FGD are installed at a LCP the residual mercury emission is 25% of the mercury in the coal.
- A residual emission of 10% is achievable when an ESP/FGD is combined with a Selective Catalytic Reduction (SCR) for the abatement of NO_x.
- The reduction rate when firing sub-bituminous coal or lignite is considerably lower and ranges from 30 - 70 %. The lower reduction rate is caused by the low amount of unburned carbon on the fly ash resulting in lower adsorption of elemental mercury

If coal fired power plants apply the most effective techniques to reduce emissions of SO_2, dust and NO_x, the emissions of mercury will also be reduced. These techniques can be regarded as Best Available Techniques (BAT) BAT and for EU Member States application of BAT is mandatory under the IPPC Directive.

In the current situation in the EU still many large combustion plants do not apply the full range of abatement techniques, in particular SCR is not common yet. Information about the number of plants that apply FGD and SCR will give a better picture of the mercury emission reduction that can be achieved in Europe. The six coal fired power plants in the Netherlands all use ESP, SCR and FGD. For three new coal fired power plants that are constructed in The Netherlands emissions of mercury are expected to be below 3 microgram/ Nm^3, as a yearly average at 6% O2 (ref. environmental permits RWE, Electrabel and E/ON). These installations apply FGD and SCR and a fabric filter, but no specific mercury abatement techniques.

In the USA about one-third of US power generating capacity was equipped with SO_2 scrubbers in 2005. The US EPA expects that in 2015 two-thirds of the installations will be equipped with FGD (ref. 16).

Specific abatement techniques for mercury reduction

At the Power Plant Air Pollutant Control "Mega" Symposium in 2008 several new methods have been presented for the reduction of mercury emissions up to removal efficiency of 90%.

Activated Coal Injection (ACI) upstream of the ESP/FF increases the Hg(P) portion that can be removed by the ESP/FF from the flue gas. When brominated active coal is injected the Hg(2+) portion also increases which improves the performance of the FGD for the removal of mercury (ref 4). The use of activated coal as adsorbent will reduce the emission of mercury to levels in the range of microgram/ Nm^3. For the Toxecon process, based on injection of activated carbon, emission levels were established in the range of 0.5 to 3 microgram/ Nm^3 (Ref. 17).

Besides activated coal other adsorbents are commercially available that will increase the Hg(P) portion and improve the emission reduction rate of the ESP/FF. (ref. 5 , 6, 7, 8).

The use of activated coal can have a negative impact on the quality of by-products of the flue gas cleaning, such as fly ash and gypsum. Instead of the injection of adsorbents fixed structures of adsorbents can be used for mercury removal. The general concept of this technology is to install a fixed sorbent structure such as gold coated plates (MerCAP™), a carbon honeycomb, or a bed of sorbent material directly downstream of a particulate collection device such as an ESP. By using a fixed bed the by-products will remain uncontaminated, the sorbent can be collected and possibly regenerated, the sorbent can oxidize mercury that is not adsorbed thus creating enhanced mercury capture in a wet scrubber, and the fixed bed can capture additional fine particulate that would otherwise be emitted (ref. 9).

The oxidation of elemental mercury can be increased by injection of chloride- or bromide compounds upstream of the SCR. Thus the FGD emission reduction rate will also be increased. This technique can also be combined with sorbent injection (ref. 10, 11, 12). Several suppliers offer brominated products that will reduce mercury emissions, e.g. Geobrom offered by Chemtura and the Vosteen process (ref. 18, 19).

When coal with low chlorine content is combusted, the amount of elemental mercury that is oxidized on the SCR catalyst is small and therefore the emission reduction rate may be insufficient. In those cases coal blending with high chlorine coal types is a cost effective measure to reduce the mercury emission (ref. 13).

Conclusion on available techniques for reduction of mercury

If a coal fired power plant is equipped with techniques to abate emissions of PM, SO_2 and NO_x the emissions of mercury will be reduced to levels below 30 microgram/ Nm^3. If the abatement techniques on a new CFPP are optimized for reduction of mercury the emission concentration can be lowered to levels of about 3 microgram/ Nm^3. In other cases mercury emissions can be abated by using available techniques based on the injection of a sorbent, e.g. activated carbon. New abatement techniques based on the use of bromine are currently being developed and tested an will become available soon.

Costs and cost effectiveness of mercury abatement

In general mercury abatement is expensive because of the low mercury concentrations in off-gases and the volatility of metallic mercury, Hg(0). This also goes for mercury abatement at CFPP.

Costs per installation and cost effectiveness

In a new coal fired power plant, equipped with SCR, FGD and fabric filters the emissions of mercury will be reduced without additional costs for mercury abatement. Reduction efficiencies depend on the input of mercury and on the design and operation of the installation, but can go up to 90%.

If additional measures are applied to reduce mercury emissions the costs depend on the situation. The most used technique is based on the injection of activated coal. If the installation is already equipped with a fabric filter this will be a relatively economical solution. A value of 11000 $/lbs Hg, about 19000 euro/kg, was reported for the Toxecon process (ref. 17).

If a fabric filter is added to the power plant only for reduction of mercury emissions the costs of the filter will be apportioned only to the mercury reduction. This will lead to a less favourable cost effectiveness. If a power plant has no emission abatement techniques installed the costs of mercury reduction will be relatively high. The total costs of a fabric filter and a system for injection of activated carbon based on the Toxecon process were estimated to be 62000 $/lbs, or 106000 euro/kg mercury abated (ref. 17).

DOE/NETL estimated the cost effectiveness of mercury abatement to range from 3810 to 166000 $/lbs Hg, or 6500 to 300000 euro/kg mercury (ref. 20). The lower values in the range are costs for installations that are already equipped with abatement techniques. In that case the costs of abatement mainly consist of the costs of the used sorbent. The high values are for installations that have to add an additional fabric filter to remove the sorbent from the off-gases.

In 2006 TNO estimated the cost effectiveness of additional specific mercury abatement on coal fired power plants at 128000 euro/kg mercury removed (ref. 21).

It is assumed that in the next 15 years all CFPP in Europe will be equipped with abatement techniques that will remove NO_x, SO_2 and PM from the off-gases. This is based on the ELVs in the new Industrial Emissions Directive in the EU and on the upcoming revision of the Gothenburg

Protocol. It can be expected that these installations will meet an ELV for mercury of 30 microgram/ Nm^3 without additional sorbent injection to remove mercury.

If the ELV for mercury in the new HM Protocol will be set at a lower level, e.g. 3 microgram/ Nm^3, additional measures may be necessary for a part of the installations. The cost effectiveness of these measures is estimated to be 10000 euro/kg mercury, based on the use of sorbent.

As a comparison the cost effectiveness of the reduction of NO_x by using an SCR is about 5 euro/kg. Reduction of SO_2 by using FGD costs less than 5 euro/kg.

The emission of mercury for a large, 1000 MW power plant is estimated to be in the magnitude of 100 kg/year. If reduction of 1 kg of mercury costs 10000 euro the additional yearly costs for abatement of mercury emissions are in the range of 1 million euros. These costs of abatement of PM and of SO_2 are estimated to be also in the range of one million euro, per pollutant, for a large CFPP. Costs for reduction of NO_x are estimated to be 2 or 3 times higher. Additional measures to reduce mercury emissions could lead to a 10 to 20% increase of the costs of emission abatement on a large CFPP. It is expected that new abatement techniques, based on the use of brominated compounds, will be more economical.

A modern 1000 MW power plant (electrical power) can produce about 7000 to 8000 GWh of electrical energy yearly. Additional mercury abatement can lead to an increase of the price of electric power of about 0.0001 euro / kWh.

It is important to notice that adding an adsorbent injection to a coal fired power plant will have several co-benefits next to mercury reduction. Other pollutants, like heavy metals and POPs, will also be reduced.

Total costs

If the total mercury emissions from CFPP in the EU are 21.2 tonnes, as reported in EPER, total costs for abatement will be 212 million euro's if all installations meet the ELV of the IED. Costs for the non-EU European countries, that will have to meet the ELV in the new Gothenburg Protocol, will be in the same order of magnitude, so that the total additional yearly costs of additional mercury abatement on CFPP in the UN ECE Europe region are estimated to be in the range of 500 million euro. If emissions are higher than reported to E PRTR, being in the magnitude of the TNO estimate at 119 ton/year, the total costs will be in the magnitude of 1000 million euro yearly.

Current policies and legislation in the UN ECE region

The USA has issued the Clean Air Mercury Rule (CAMR) in 2006, aimed at reducing mercury emissions from power plants from 48 t/a in 2005 to 15 t/a in 2018.

In the EU the LCP directive (2001/80/EC) has no specific ELVs for mercury. The LCP Directive will become part of the new Industrial Emissions Directive (IED). The draft of the IED does not give ELVs for mercury emissions, but it gives ELVs for SO_2, NO_x and PM. For the large power plants these ELVs are 200, 200 and 20 mg/ Nm^3. To meet these ELVs specific abatement techniques have to be applied for at least the reduction of SO_2 and dust. For new installations the ELVs will be set at 150, 150 and 10 mg/ Nm^3. This is more or less on the same level as the proposals for the option 2 for revision of the Gothenburg Protocol. These emission levels imply the use of additional NO_x reduction. This will lead to reductions of mercury emissions with an efficiency of about 75%, or about 90% if an SCR is used, as explained above. The remaining emissions of mercury after emission abatement will be in the range of several microgram/Nm^3.

Research in China has shown that mercury emissions from coal fired power plants, equipped with ESP, FGD and sometimes SCR can range from 2 to 27 microgram/ Nm^3 (ref 22).

It is unclear whether individual EU Member States have legislation or policies in place aimed at mercury reduction from CFPP. Germany has announced a proposal for an ELV of 3 microgram/ Nm^3 (ref. 23). In environmental permits for three new installations in The Netherlands the ELVs are set at 2.4 or 2.8 microgram/ Nm^3 as yearly average (ref. 24).

Proposal for revision of the HM Protocol (ECE/EB.AIR/WG.5/2010/10)

In the proposal to the WGSR, ECE/EB.AIR/WG.5/2010/10, in chapter G, paragraph 12, an ELV for mercury from power plants is proposed of 30 microgram/ Nm^3. This ELV can be met by applying abatement techniques that are already mandatory to meet the new European Emission Directive IED. These techniques are also necessary to meet the expected ELVs in the revised Gothenburg Protocol, when option 1 or option 2 for the revised ELVs is applied.

Further reduction of mercury emissions can be accomplished by applying lower ELVs for mercury, e.g. a value of 3 microgram/ Nm^3. If abatement techniques are designed to meet also the ELV for mercury this will not lead to higher costs. If an existing installation, applying SCR, FGD and a fabric filter, does not meet an ELV of 3 microgram/ Nm^3 additional measures in the shape of injection of a sorbent or addition of a halogenated compound is necessary. The costs of these measures will be relatively low compared to costs of mercury abatement at other stationary sources like steel plants or cement kilns.

Conclusion

Coal fired power plants are by far the largest source of mercury to air. When coal fired power plants meet the emission limit values for NO_x, SO_2 and PM in the European Industrial Emissions Directive or the emission limit values according to option 2 in the proposal for revision of the Gothenburg Protocol, the emissions of mercury will be reduced to concentration levels below 30 microgram/ Nm^3. No additional measures are required to meet the ELV that is proposed for the revised HM Protocol. Emissions from coal fired power plants would then still emit around 50% of the total emission in Europe.

BAT to reduce mercury emissions is the application of sorbent injection , which can reduce emission concentrations by 90%. Thus an emission limit value for coal fired power plants can be reached of 3 microgram/Nm^3.

The costs of additional sorbent injection on coal fired power plants that meet the emission standards of 3 microgram are estimated to be in the magnitude of 10.000 euro/kg mercury abated. For a large power plant of 1000 mW the costs would be 1 million euro or 0.0001 euro/kWh. For the whole European UN/ECE region the costs are calculated to be 500 million euro.

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