Journal of the Air & Waste Management Association

ISSN: 1096-2247 (Print) (Online) Journal homepage: http://www.tandfonline.com/loi/uawm20

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To cite this article: Ruud Meij , Leo H.J. Vredenbregt & Henk te Winkel (2002) The Fate and Behavior of Mercury in Coal-Fired Power Plants, Journal of the Air & Waste Management Association, 52:8, 912-917, DOI: 10.1080/10473289.2002.10470833

To link to this article: http://dx.doi.org/10.1080/10473289.2002.10470833

Published online: 27 Dec 2011.

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TECHNICAL PAPER

ISSN 1047-3289 *J. Air & Waste Manage. Assoc.* **52**:912-917
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The Fate and Behavior of Mercury in Coal-Fired Power Plants

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ABSTRACT

For the past 22 years in the Netherlands, the behavior of Hg in coal-fired power plants has been studied extensively. Coal from all over the world is fired in Dutch power stations. First, the Hg concentrations in these coals were measured. Second, the fate of the Hg during combustion was established by performing mass balance studies. On average, 43 ± 30% of the Hg was present in the flue gases downstream of the electrostatic precipitator (ESP; dust collector). In individual cases, this figure can vary between 1 and 100%. Important parameters are the Cl content of the fuel and the flue gas temperature in the ESP. On average, 54 ± 24% of the gaseous Hg was removed in the wet flue-gas desulfurization (FGD) systems, which are present at all Dutch coal-power stations. In individual cases, this removal can vary between 8% (outlier) and 72%.

On average, the fate of Hg entering the power station in the coal was as follows: <1% in the bottom ash, 49% in the pulverized fuel ash (ash collected in the ESP), 16.6% in the FGD gypsum, 9% in the sludge of the wastewater treatment plant, 0.04% in the effluent of the wastewater treatment plant, 0.07% in fly dust (leaving the stack), and 25% as gaseous Hg in the flue gases and emitted into the air. The distribution of Hg over the streams leaving the FGD depends strongly on the installation. On average, 75% of the Hg was removed, and the final concentration of Hg in the emitted flue gases of the Dutch power stations was only ~3 $\mu g/m_{STP}^3$ at 6% O_2. During co-combustion with biomass, the removal of Hg was similar to that during 100% coal firing.

Speciation of Hg is a very important factor. An oxidized form ($HgCl_2$) favors a high degree of removal. The conversion from Hg^0 to $HgCl_2$ is positively correlated with the Cl content of the fuel. A catalytic DENOX (SCR) favors the formation of oxidized Hg, and, in combination with a wet FGD, the total removal can be as high as 90%.

IMPLICATIONS

The U.S. Environmental Protection Agency (EPA) recently decided that regulation of Hg emissions from coal-fired electric power plants is necessary and appropriate. In response to this determination, this paper presents the experiences of the last 20 years in the Netherlands.

INTRODUCTION

About 40% of the centrally generated electricity in the Netherlands is generated from coal. Only imported bituminous coal is fired in pulverized dry-bottom boilers equipped with high-efficiency electrostatic precipitators (ESP) and wet flue-gas desulfurization plants (FGD). The release of trace elements is an important environmental issue. In the past decade, extensive measurements have been performed at Dutch coal-fired power plants to establish the fate of the trace elements.[1] These measurements were carried out by KEMA Power Generation & Sustainables. This paper will focus on the results concerning Hg.

Most trace elements end up in the ash, ~99.9% of which is collected as bottom ash and as pulverized fuel ash (PFA; ash collected in the ESP). Accordingly, the emissions are very low.[1] For Hg, however, the situation is different. During combustion, the Hg in the coal is released as gaseous elemental Hg. However, the largest part by far, ~75%, is removed in the combined flue gas systems (ESP and FGD). The adverse effect is that ~25% of the Hg remains in the vapor phase in the flue gases and is released into the air.

In this paper, the amount of Hg introduced by coal and the fate of the Hg at the power plant, including removal in the FGD, is quantified. This is an update of a previous article from 10 years ago.[2] Since then, more data have become available, especially from co-combustion of secondary fuels such as biomass and waste. The Hg emissions of Dutch power plants and their effect on the ambient air concentrations and local depositions are discussed in a separate paper.[3]

EXPERIMENTAL

Flue gases for Hg analyses were sampled by different methods.[4] The first method is using activated carbon. Flue gases were sampled at a flow of 1 L/min and were led over activated carbon in a cartridge with two compartments, each with a diameter of 10 mm and a length of 50 mm, at a temperature of ~60 °C. Aerosols were separated by a plane Teflon filter. Spherical activated carbon (KUREHA) was used (grade: BAC MP, diameter 0.1–1 mm). The second method is according to NEN-EN 13506, using "De Graaff" wash bottles with a content of ~100 mL. Flue gases were sampled at a flow of 1–2 L/min through two wash bottles containing 4% $K_2Cr_2O_7$ in 4-M HNO_3, or 0.6% $KMnO_4$ in 2-M H_2SO_4. These two wash bottles were preceded by a

wash bottle containing 2% H_2O_2 to remove SO_2. If speciation of Hg was measured, a fourth wash bottle, filled with 1-M HCl, was used as the second one for sampling Hg^{2+}; the last two bottles were then used for sampling of Hg^0.

The solid samples were digested using HNO_3–HF at 180 °C in a pressurized Teflon-lined decomposition vessel under O_2 atmosphere, according to U.S. standard ASTM-D3684 or by Dutch standard NVN 2507. Atomic fluorescence spectroscopy (AFS) was used for chemical analysis. The activated carbon was analyzed by instrumental neutron-activation analyses (INAA). In some coal and ash samples with very low Hg concentrations, the following procedure was applied to obtain results with an accuracy of ±1 µg/kg. After irradiation with neutrons, a preconcentration was performed, whereby the samples were heated in a quartz tube in a nitrogen atmosphere (<950 °C) and the Hg vapor, collected on activated carbon, was measured with gamma spectrometry.[5]

Table 1. Mercury concentrations in bituminous steam coal as imported in the Netherlands and analyzed by KEMA.[a]

mg/kg	*n*	Mean		*s*	*v* (%)
Australia	17	0.08	±	0.06	70
Colombia	7	0.06	±	0.03	57
China	2	0.15			
Egypt	1	0.10			
Germany (Ruhr area)	1	0.16			
Indonesia	7	0.04	±	0.02	63
New Zealand	1	0.05			
Norway (Spitsbergen)	2	0.14			
Poland	10	0.35	±	0.55	154
Russia (Kuzbass)	1	0.06			
South Africa	12	0.09	±	0.02	23
United States (eastern)	15	0.14	±	0.12	84
Venezuela	2	0.08			
Blend	36	0.09	±	0.07	72
Total	109	0.12	±	0.19	161
Weighted averaged in the Netherlands for 1999		0.11	±	0.2	174

[a] *n* is number; *s* is standard deviation; *v* is coefficient of variation ($= \frac{s * 100}{\text{mean}}$).

RESULTS AND DISCUSSION

Mercury Content of Coal

The results for 109 coal samples, which were analyzed for Hg, are presented in Table 1. The lowest concentrations were found in coal from Australia, Colombia, and Russia. Higher concentrations were found in coal imported from the eastern part of the United States, Spitsbergen, China, and Germany. The highest concentrations were found in coal imported from Poland. In general, the concentrations are low (<0.1 mg/kg) and the range is narrow (0.03–0.3 mg/kg). The average value is 0.12 mg/kg. For 1981–1982, a weighted average on the origin of the coal fired in the Netherlands is 0.15 mg/kg; for 1990, it is 0.12 mg/kg; and for 1999, it is 0.11 mg/kg. The figures for U.S. coal are in agreement with the median value for eastern U.S. coal, as given by the Illinois State Geological Survey (0.17 mg/kg).[6,7]

Many samples were taken during mass balance studies, in which case sampling took place after milling, so as to obtain homogeneous samples. Homogeneity was achieved by the smaller particle size (<100 µm), the removal of large fragments consisting of pyrite from the raw coal in the mills, and the grinding process. Also, more homogeneous samples were obtained at the power plant as compared with the mine samples. This is a result of the various handling processes during transportation from the mine by train, sea vessel, and river vessel to the power plant, the stacking processes, and, finally, the reclaiming processes at the trench bunker of the power plant. This is also illustrated by the spread of the Hg concentrations in one lot or blend (see Table 2). In these cases, samples were taken for 3–5 days. In 2000, during 7 weeks, samples were taken from a blend made of five different coals. For a trace element in concentrations less than 1 ppm, the absolute standard deviation is remarkably low. They are comparable to results obtained by inter- and intralaboratory investigations of Hg in coal,[8] suggesting that the deviations in Table 2 are of analytical origin.

Table 2. The dispersion of Hg in one lot or blend of bituminous coal.

mg/kg		*n*	Mean		*s*	*v* (%)
Poland	1982	3	0.20	±	0.02	8
United States (eastern)	1983	10	0.08	±	0.02	28
United States (eastern, blend of two lots)	1986	3	0.27	±	0.02	6
Poland	1998	3	0.077	±	0.003	4
Blend (raw coal)	2000	14	0.094	±	0.027	29

Mercury Content of Coal Ash

To describe the enrichment of Hg in the ash with respect to the coal, the term "relative enrichment" (RE) is used:[1,9]

$$RE_{\text{Hg}} = \frac{[Hg]_{\text{ash}}}{[Hg]_{\text{coal}}} * \frac{\text{ash content of the coal in \%}}{100} \quad (1)$$

This means that if the RE factor for a certain element is equal to 1, the entire element originally present in the coal is concentrated in the ash that remains after combustion. The RE factors of Hg in the bottom ash and the ash as

collected by the ESP, called pulverized fuel ash (PFA) in the United Kingdom and fly ash in the United States, are given in Table 3. The RE factor in the bottom ash is very low (<0.1), and the RE factor of the PFA is ~0.6. It appears that Hg is strongly depleted in the bottom ash and moderately depleted in the PFA. The coarse ash particles (>100 µm) rather quickly fall to the bottom of the boiler and are called bottom ash or slag. Because contact with the flue gases is limited, the concentration of Hg in the bottom ash is very low and mostly below the limit of detection. In two series, the exact concentration can be established (RE = 0.1), and ~2% of the Hg originally present in the coal is found in the bottom ash. On average, ~50% of the Hg originally present in the coal is found in the ash, mainly the PFA.

Mercury Concentrations in the Flue Gases Downstream of the ESP

The measurements of Hg in the flue gases downstream of the ESP performed by KEMA since 1980 are given in Table 4. The corresponding vaporization percentage with respect to the concentration in the coal is also given. On average, 43% of the Hg present in the coal is found in the flue gases in the vapor phase. This is in agreement with the findings in the ashes, so the mass balance seems to be correct. The contribution of aerosol-bound Hg is negligible because the particulate loadings are low, as are the Hg concentrations in the fly ash particles.

The vaporization percentage of Hg downstream of the ESP can vary. Regardless of the form in which Hg is present in the coal, elemental Hg is released during combustion. Elemental Hg is very volatile, and, accordingly, little will be removed in the ESP. The vaporization percentage can be expected to be ~90%. However, in the presence of HCl, Hg^0 can be (partly) converted into $HgCl_2$ at temperatures less than 800–500 °C, for example, by the following reactions:[1,2,10,11]

$$Hg + 2HCl \rightarrow HgCl_2 + H_2 \quad (2)$$

$$Hg + 4HCl + O_2 \rightarrow 2HgCl_2 + 2H_2O \quad (3)$$

Hall et al. demonstrated the occurrence of such reactions under laboratory conditions.[10] Also, thermodynamic calculations indicate the possible presence of $HgCl_2$, which is less volatile and begins to condense on the fly-ash particles if the temperature in the ESP is not too high (less than 140 °C). Besides that, $HgCl_2$ is rather sticky, so if the temperature is favorable, it adsorbs onto ash particles. Lower vaporization percentages of Hg are found when the concentrations of HCl in the flue gases are relatively high, and, in that case, relatively more Hg is found in the PFA.[2]

For 28 test series in the Netherlands, the relationship between the chlorine content of the coal and the Hg vaporization percentage is presented in Figure 1. A similar relation to that found under laboratory conditions[10] is thus found in practice. Only the results of Test Series XII and XVI deviate. In the particular case of Test Series XVI, it was found that ~53% of the Hg was present in a water-soluble form, most probably as $HgCl_2$. However, because of the high temperature of the flue gases (140–150 °C), $HgCl_2$ is still present in the vapor phase. The results of Test Series XII also deviate, most probably due to flue gas temperatures of greater than 140 °C. The normal operating temperature of the ESPs in the Netherlands is ~120 °C. Only the test series indicated with a star in Figure 1 are the ones for which complete mass balances of Hg were measured. All those mass balance studies gave satisfactory results (mean 0.96 ± 0.23)[2] (see also Table 7).

Measurements of the speciation of Hg in the flue gases can give more insight. In the last 5 years, this has become available routinely. On the basis of literature data and KEMA measurements, a graph has been made of the conversion of Hg^0 to Hg^{2+} as a function of the Cl content in the coal (see

Table 3. Concentrations and relative enrichment factor of Hg in bottom ash and pulverized fuel ash (PFA = ESP – ash).

	n	Mean		*s*	*v* (%)
Concentrations in mg/kg					
Bottom ash	15	<0.3			
PFA (ESP – ash)	29	0.28	±	0.13	46
RE Factor					
Bottom ash	9	<0.1			
PFA (ESP – ash)	19	0.6	±	0.29	47

Table 4. Statistical data on the concentration and vaporization percentages of gaseous Hg in flue gas downstream of the ESP.

n	Low	High	Mean	*s*	*v* (%)
Concentrations in the Flue Gases Downstream of ESP in µg/m³					
37	0.3	35	4.9	7.3	149
Vaporization Percentage Relative to the Concentration in Coal					
36	1.0	115	43.4	30.1	69

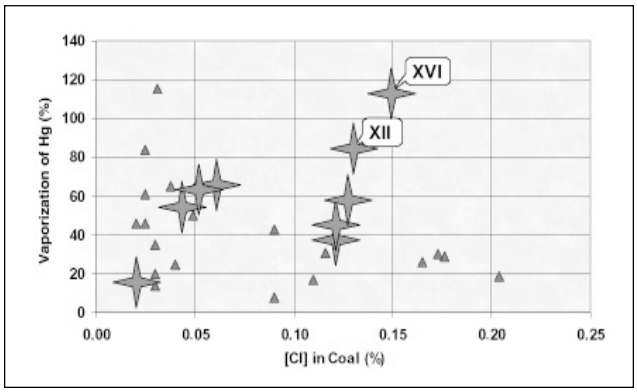

Figure 1. The relation between vaporization percentage of Hg and the Cl content of the coal. The points indicated with a star refer to complete mass balance studies.

Figure 2).[12,13] Figures 1 and 2 form each other's mirror image.

Our results deviate from measurements done by Devito and Rosenhoover in the United States.[14] They found very little Hg condensation on the PFA. Also, in Germany, the removal of Hg in ESPs is lower (6 and 17% removal,[11] and 26%[15]). The reason is that the ESPs studied in the United States are operated at a higher temperature than those in the Netherlands. This is because of the higher sulfur concentrations in the coal, which require higher temperatures in the ESPs to prevent condensation of H_2SO_4.

Gutberlet et al. have studied the speciation of Hg in the flue gas in detail both upstream and downstream of an SCR installation.[11] It was demonstrated that both elemental Hg and $HgCl_2$ were present in the flue gases. The fraction of elemental Hg for various coals and boilers (dry-bottom and slag-tap) lies between 40 and 60% upstream of a high-dust SCR installation at temperatures of ~380 °C. After the SCR installation, the proportion of metallic Hg falls between 2 and 12%. These results are confirmed by measurements performed by KEMA at similar Dutch installations. It can be concluded that elemental Hg is oxidized to Hg^{2+} in the SCR installation. This means that the ESP and FGD plant might remove a larger part of the Hg.

Mercury Concentrations in the Flue Gases Downstream of the FGD Plants

Flue-gas desulfurization systems are installed primarily to remove SO_2. However, other flue gas components also can be removed in this process. An overview of the measurements of Hg in the flue gases downstream of a wet FGD plant, performed by KEMA since 1986, is presented in Table 5. The corresponding removal percentage, with respect to the concentration in the flue gases upstream of the FGD (downstream of the ESP), is also given in Table 5.

Just as the removal in the ESP is dominated by the speciation of Hg, this is also true for the removal by the FGD plant. Wet scrubbers effectively remove $HgCl_2$, but Hg^0 is barely removed. Besides, liquid redox reactions, by which $HgCl_2$ can be (re)converted to Hg^0, can occur in the scrubber. In addition, adsorption of Hg onto walls occurs. In a steady-state environment, the adsorption and desorption will be in equilibrium. But in a dynamic situation, such as changes in flow, temperature, or Hg concentration in the flue gases (changes in fuel), this equilibrium can be distorted. It is thus possible that, on a short time period, it seems that more Hg is leaving the FGD than is entering. Establishing equilibrium can take many days. However, on average, more than 50% of the Hg is removed in a wet FGD. These results are in agreement with the findings of others: Devito and Rosenhoover[14] in the United States; Curtis[16] in Canada; Yokoyama[17] in Japan; Maier et al.[18] in Germany and Austria; and Sander[19] in Denmark.

It has to be mentioned that Hg removal in a wet FGD cannot be considered independently, because it is influenced by the other flue gas cleaning systems. If $HgCl_2$ is already effectively removed in an ESP, the removal from the next FGD will be lower. In the end, their combined removal is improved.

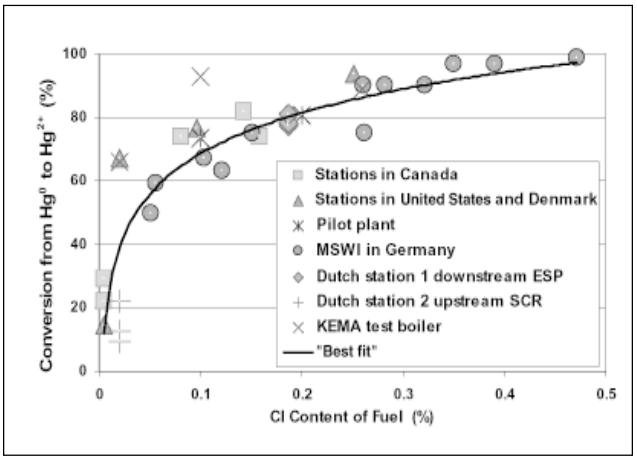

Figure 2. Conversion from Hg^0 to Hg^{2+} as a function of the Cl content of the fuel.

Mercury Concentrations in the Streams around an FGD Plant

In an FGD plant, more streams are involved. The input streams are limestone or lime and process water. The outgoing streams are gypsum and wastewater. The wastewater is cleaned in a wastewater treatment (WWT) plant, which produces effluent and sludge. The Hg content of these streams is given in Table 6. The concentrations of the solid samples are given on a dry basis.

Mass Balances

Three types of mass balances are made. One mass balance is made around the boiler and ESP; one around the FGD, including the WWT plant; and one around the whole installation. The first one is only applicable to stations without FGD and is presented in Table 7. The mass balances vary between 70 and 180% and average 96%. The variations between the different test series have been previously discussed, in the section titled "Hg Concentrations in the Flue Gases Downstream of the ESP" and in Figure 1. The results from Station 2 differ from the other results. This is because of the presence of an SCR, by which Hg^0 is transformed into Hg^{2+} and is then adsorbed on the ash particles.

Table 5. Statistical data on the concentration and removal percentages of gaseous Hg in FGD plants.

n	Low	High	Mean	*s*	*v* (%)
Concentrations in the Flue Gases Downstream of FGD in μg/m³					
5	0.6	2.4	1.3	0.71	53
Removal Percentage in FGD					
6	8[a]	72	54	24	44

[a]This figure deviates from the other measurements and is probably an outlier.

Table 6. Mercury content of streams around an FGD plant.

Ingoing Streams			Outgoing Streams		
Limestone	mg/kg	<0.2–<0.5	Gypsum	mg/kg	0.058–0.92
Lime	mg/kg	0.01–<0.5	Effluent	mg/L	0.4–10
Process water	µg/L	<1–2	Sludge	mg/kg	4–26

The figures used in the KEMA TRACE model, which predicts the emissions and ash composition for an average power station, are given in the lowest part of the table. They differ from the average results, because the model is based on a larger database of RE factors of Hg in the ash (see section "Hg Concentrations in the Ash" and Table 3).

Setting up mass balances around the FGD and the WWT plant is much more complicated, because many parameters are involved and the situation is very dynamic. Results from three studies are given in Table 8. The results of Test Series XII are discussed in detail in ref 20.

The low balance value found for Station 1 is the result of a not yet reached equilibrium. Twenty hours before the measurements, the Hg input via the fuel was increased, resulting in increasing Hg concentrations in the wash suspension of the FGD during the measurements. Furthermore, one has to keep in mind that it concerns a trace element. The unbalance is only 6 g Hg/hr, which can easily accumulate in the wash suspension of ~1500 m^3 in the FGD vessel. The unbalance of Station 2 is only 4 g Hg/hr. The contribution of the FGD and the WWT plant to the total Hg mass balance is only 20%, which means that the total mass balance of Hg at Station 2 is 109%. Table 8 illustrates that the partitioning of Hg between the sludge and gypsum is very much dependent on the type of installation. The total mass balance of the whole installation is presented in Figure 3. The average emissions into the air of 25% are in agreement with a recent study at a German power station equipped with DENOX.[15]

Influence of Cofiring

In daily practice, the sludge of the wastewater treatment plant is cofired with the coal. This means that Hg present in the sludge is reintroduced to the boiler and follows the same route again. In the end, the Hg concentrations in all the other streams are increased by ~10%, and most of it ends up in the ashes and gypsum. The concentrations in the ash and gypsum are low and cause no environmental problem.

To reduce the CO_2 emissions of the long carbon cycle, in the Netherlands, practically all coal-fired stations cofire secondary fuels nowadays, such as biomass (bone and meat meal, waste wood, paper sludge, etc.). At KEMA, the fate of trace elements, such as Hg, also has been well studied for cofiring. Beginning in 1993, eight test runs were performed in the 1-MW_{th} KEMA test boiler, followed by 10 measurements on real scale at seven different power stations. In all cases, the percentages of cofiring on dry mass basis were less than 10%. It appears that the behavior of the elements, including Hg, is the same as with 100% coal firing. Thus, all the RE factors, vaporization percentage, and removal in the FGD are also applicable to cofiring up to 10 mass %. At present, in the KEMA test boiler, a new test series is being performed at higher percentages of cofiring.

Table 7. Mass balances of Hg around a boiler and an ESP, partition in %.[a]

No.	Origin Coal	Bottom Ash	PFA	Flue Gases	Out/In (%)
IV	Australia	1	8	91	80
XI	United States	< 1	41	59	70
XII	United States	< 1	10	90	90
XIIIb	Australia	< 1	43	57	120
XIV	Colombia	< 1	27	73	80
XVI	United States	0.2	6	94	120
Station 1	Poland	< 1	54	46	130
Station 2	Blend	< 2	77	21	80
	Mean	< 1	33	65	96 ± 23
Averaged figure as used in the model					
	Mean	< 1	49	50	100

[a]Normalized to 100%.

CONCLUSIONS

In the Netherlands, the fate of Hg entering a coal-fired power station has been well studied since 1980. For the modern coal-fired power stations in the Netherlands, equipped with high efficiency ESPs and wet FGDs, it appears that, on average, 50% of the Hg is removed in the ESP via the PFA (ash collected in the ESP). The remaining Hg is, on average, removed by 50% in the FGD plant. This means that, on average, 25% of the Hg present in the fuel is emitted into the air. The Hg concentrations in the coal, as fired in the Netherlands, are very low, on average 0.11 mg/kg, resulting in low flue gas concentrations in the stack of ~3 $\mu g/m_{STD}^3$.

The speciation of Hg is a very important parameter and explains the variations between the different measurements.

Table 8. Mass balances of Hg around an FGD and a WWT plant, partition in %.[a]

	In Process Water	Flue Gases	Lime-stone	Out Flue Gases	Gypsum	Sludge	Effluent	Out/In %
XII	>2	>66	<31	>23	<62	>9	>6	90
Station 1	n.m.	100	n.m.	88	9	4	0.02	40
Station 2	<0.2	86	14	16	75	8	0.2	250
Model[b]	0.5	99	0.5	50	33	17	<0.1	100

[a]Normalized to 100%; [b]Averaged figure as used in the model.

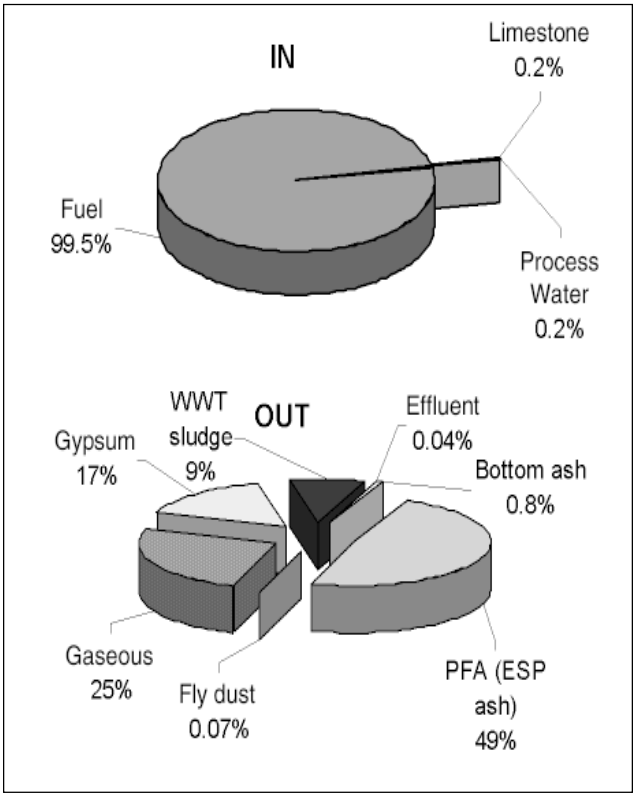

Figure 3. Modeled mass balance of Hg in a typical Dutch coal-fired power station equipped with ESP and wet FGD.

Hg can be present as Hg^0 and as $HgCl_2$. Elemental Hg is difficult to remove, while oxidized Hg is more easily removed. The speciation is influenced by the presence of HCl in the flue gases. Also, the presence of a DENOX of the SCR type promotes the oxidation of Hg. The operating temperature of the ESP also influences the removal of Hg; lower temperatures promote higher removals. During cofiring with sludges and biomass, studied with up to 10% (m/m) cofired material only, the mechanisms are the same as for 100% coal firing.

In an FGD, the removed Hg ends up mainly in the sludge and gypsum. The partition between these two streams depends strongly on the operating conditions of the installation. The emission of Hg via the effluent of the wastewater treatment plant is negligible.

ACKNOWLEDGMENTS

The project described in this paper was carried out within the framework of the Collective Research Order placed with KEMA by the four electricity-generating companies in the Netherlands: Electrabel Nederland N.V., E.ON Benelux, Essent Energie Productie, and Reliant Energy Europe Power Generation Benelux.

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