Comparison of Material Flows in Sewage-free and Sewage-generating Flue-Gas Purification Systems of Municipal Waste Incineration Plants

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Abstract

During incineration of wastes in waste incineration plants, polluted flue gases are generated which have to be subjected to flue gas purification. Although the legal requirements are nearly unambiguous, the question of whether wet flue gas purification is to be performed in a sewage-free or sewage-generating manner is discussed controversially by experts in the Federal Republic of Germany.

As a contribution to this discussion, material flow studies of sewage-free and sewage-generating flue gas purification processes in waste incineration plants were performed by ITAS in cooperation with ITC-TAB. The study covered three waste incineration plants, two of which were operated in a sewage-generating and one in a sewage-free manner.

The results obtained can be summarized as follows:

The data and information submitted by most of the plant operators are not sufficient for a comprehensive balancing of flue gas purification systems in waste incineration plants. For this reason, plant operation often is not optimally tailored to the substances prevailing. During operation, at least temporary strong superstoichiometric dosage of auxiliary chemicals cannot be excluded. By means of plausibility assumptions and model calculations, closed balancing of most plants could be achieved.

Moreover, it was demonstrated by the balancing of technical-scale waste incineration plants that the material flows in "wet" flue gas purification are less dependent on the design of the flue gas purification section (sewage-free/sewage-generating), but considerably affected by the operation of the flue gas purification system (e.g. volume of absorption agents used). Hence, material flows can be controlled in a certain range.

Introduction

Thermal waste treatment generates flue gases which must be cleaned of pollutants before being released into the atmosphere. A legal regulation (17th German Federal Environmental Impact Control Ordinance) issued in 1990 defined new limits for municipal solid waste incinerators (MSWI) which are below the corresponding requirements of power plants.

In flue gas purification devices, a dust removal system may be followed by multistage scrubbers in which the pollutants are absorbed by scrubbing liquids. The scrubbing liquids can either be evaporated internally or passed to a sewage treatment plant. As a consequence of relatively high heavy metal concentrations in the waste water of MSWI detected in the seventies, the possibility of the release of waste water from MSWI should be excluded in general. Waste water from MSWI may no longer be passed to sewage treatment plants in Germany since the late eighties. Although the legal requirements are nearly unambiguous, the question of whether wet flue gas purification is to be performed in a

sewage-free or sewage-generating manner is discussed controversially by experts in the Federal Republic of Germany. Arguments used in favour of discharging such waste waters are the lower pollutant loads arising from modern waste incineration plants, and the added processing expense in sewage-free operations.

In this study, material flow analyses are established for sewage-generating and sewage-free flue gas purification systems of technical MSWI. Material flow analyses are a tool used to trace and make transparent the use and whereabouts of specific substances, by type, volume and mass, with all ramifications in the process taken into account. The basic data of these material flow analyses were conducted from informations received from technical-scale MSWI.

Approach

Three technical-scale MSWI were investigated. All plants, for which balances were established, have electrostatic precipitators for dust removal downstream of the boiler, followed by two stage scrubbing systems. One of the plants selected is equipped with a sewage-free flue gas purification system. The others discharge their waste water into a municipal sewage treatment plant. Additional information was used for plausibility assessments.

The bounderies of the system selected for the material balances do not extend over the entire MSWI. The area of coverage begins with the raw gas downstream of the boiler and ends downstream of the scrubbers of the flue gas purification system. In all plants, the offgas referred to as clean gas in the figures in this study is passed through additional flue gas purification devices to remain under the limits under the 17th German Federal Environmental Impact Control Ordinance. These additional flue gas cleaning devices have no impact on the flows of the pollutants considered.

The results described here are limited to SO_2, HCl, and mercury. Conclusions are derived from a comparison of the balances and, finally, the findings are evaluated.

Description of the Plants

The plants studied were grate firing systems incinerating more than 75% of solid municipal waste and, in addition, bulky waste and sewage sludge. No detailed description of these common technical systems for waste incineration and the required flue gas purification devices will be given at this point. In all plants considered, the concentrations of the pollutants in the raw gas are on the order of magnitude typically found in German MSWI. Table 1 lists some important data about the raw gas in the plants for which balances were established. In addition to flue gas data, also extensive information was available about the use of auxiliary chemicals for neutralization and heavy metal precipitation.

Balance Plant A

There is excellent information available about Plant A, which allowed an exact balance of the plant including the waste water treatment plant. As a consequence of the greatly varying concentration data, ranges of values are presented in some of the diagrams below.

According to figure 1, 1 t of waste contains 4.2 to 10.7 kg of chlorine which passes almost entirely into the raw gas during incineration on the grate. In the first flue gas purification step, the electrostatic precipitator, the dust separation removes the chlorides bound in the filter ash. The first scrubber removes between 3 and 6.8 kg of chlorine per 1 t of waste absorbed by the scrubbing liquid. The second scrubber absorbes small quantities of chlorine, on the order of 0.2 to 0.8 kg per 1 t of waste. Only small quantities

of chlorine remain in the gas which leaves the scrubbers. Liquids removed from the scrubbers are first subjected to waste water treatment and heavy metal precipitation and then passed to the sewage treatment plant. This flow of material contains 1.2 to 5.2 kg of chlorine per 1 t of waste.

The Sankey diagram in figure 2 shows a different distribution of material flows for sulfur than for chlorine. The slag and the filter ash accommodate 1.6 to 2.1 kg/t and 0.5 to 1.1 kg/t, respectively, of the sulfur input which, in Plant A, varies between 3.1 and 4.8 kg/t. Sulfur is removed from the flue gas in the second scrubber which uses NaOH for absorption. Waste water treatment causes a rearrangement of salts, producing low soluble gypsum which must be disposed of as residue. Only very small quantities of sulfur are passed to the sewage treatment plant.

The data available indicate that 1 t of waste contains 2 g of mercury. Figure 3 shows the distribution for mercury expected to arise from the properties of mercury. In the incineration process, most of the mercury is passed to the flue gas as $HgCl_2$ and is taken up by the liquid phase in the first scrubber. After neutralization, the mercury can be removed from the aqueous phase almost completely by means of precipitating agents. In this way, some 1.5 g of mercury per ton of waste is disposed of together with the residues from the waste water treatment. Only very low concentrations are detected in the discharge pipe to the sewer system.

Balance Plant B

MSWI B, too, is equiped with a flue gas purification system which generates waste water. Detailed data were available on Plant B for the concentrations of chlorine and sulfur in the scrubber effluents. But informations about the composition of the raw gas and the residue to be deposited were missed. Moreover, only the total volume flow of the scrubber effluents were known, whereas the volume flows from the individual scrubbers were not known. As result of a modernisation, the flue gas purification system now is no longer operated in the mode shown in figure 4, figure 5 and figure 6.

A model had to be developed for balancing Plant B against the backdrop of missing data. This model, plus some plausible assumptions, allowed a balance of MSWI B and sankey diagrams were established for sulfur, chlorine, and mercury.

For chlorine, a raw gas mass flow of 5.9 kg/t of waste was calculated from the model. The sankey diagram for chlorine in figure 4 shows that also in MSWI B large quantities of chloride, some 97% of the total input, are removed from the flue gas in the first scrubber. Approx. 3.8 kg of chlorine per ton of waste is fed by effluents from the flue gas cleaning to the sewage treatment plant. The calculated material flow to the landfill amounts to 1.9 kg/t of waste, which is in the range calculated for other MSWI.

The material flows of sulfur are shown in figure 5. The diagram indicates that, according to the model calculation, incineration of 1 t of waste gives rise to 1 kg of sulfur contained in the raw gas from which the dust has been removed. The subsequent further distribution of sulfur in the material flows clearly differs from that in MSWI A. Larger quantities of sulfur are removed in the first scrubber due to larger water volumes used in the flue gas cleaning system of MSWI B. The increase in gypsum solubility due to chloride, and the large water volumes, then lead to large amounts of sulfur being passed to the sewage treatment plant.

Figure 6 shows the mercury balance of MSWI B. The data available indicate approximate 1.8 g per ton of waste input, which is a value indicating the current customary mercury content of the waste. Some 63% of the mercury inventory in the gaseous fraction is removed in the scrubbers. The remaining 37% of mercury (0.59 g/t of waste) are released. According to the available data, approximately 0.5 g/t of waste is passed into the waste water to the sewage treatment plant. The same amount is contained in the

solid to be deposited. A part of the mercury contained in the waste water from flue gas purification system is returned to the incineration plant with the sewage sludge of the sewage treatment plant. In the absence of data, no information can be presented on the mercury load of the returned treatment sludge.

Balance Plant C

In contrast to MSWI A and MSWI B, MSWI C is equipped with a wet flue gas purification system which operates in a sewage-free manner. The flue gas purification system is made up of an electrostatic precipitator, a spray dryer, a second electrostatic precipitator, scrubber 1 for HCl and mercury, and scrubber 2 for SO_2. The brines produced in the scrubbers are evaporated in the spray dryer. The scrubber 1 and the scrubber 2 are arranged in such a manner that the loaded liquid of scrubber 2 is piped to scrubber 1, which makes balancing difficult. It must also be taken into account that more waste incineration boilers are installed than flue gas purification systems. This construction causes a mix of the material, which also explains the considerable variance in measured data.

Only the data necessary for plant operation are available. The operator was able to provide information about the raw gas, the clean gas, and the composition and volume, respectively, of the residues to be landfilled. No concrete analytical results from identical measurement periods are available.

The diagrams below show the material flows in the flue gas purification systems. The two flue gas purification systems were combined for the balance of MSWI C, because of the impossibility to balance them individually. Moreover, the residue flows to be landfilled are hatched, because the raw gas input to the flue gas purification system does not agree with the calculated output to the landfill. For this reason, it was not possible to draft exact sankey diagrams.

As is shown in figure 7, the raw gas contains a chlorine load of 3.5 - 8.7 kg per ton of waste. The average chlorine mass flow is approximately 6.4 kg/t of waste. The clean gas retains only some 0.3% of the chlorine from the raw gas. Practically the whole chlorine load from the raw gas is fed to a landfill as a solid residue.

According to figure 8, the mean sulfur load in the raw gas from which the dust has been removed amounts to approximate 0.7 kg/t of waste. The clean gas still contains approximately 4 - 5% of the sulfur from the raw gas.

The mercury load in the gaseous fraction of the raw gas from which the filter ash has been removed varies between 0.3 and 3.1 g per ton of waste. Figure 9 shows that the average mercury load amounts to approximately 1 g/t of waste.

Comparison of Plants

This comparison first of all shows that the chlorine mass flows amounting to 6 or 7 kg of chlorine per ton of waste are on the same order of magnitude in the three plants for which balances have been established. The amounts of chlorine discharged into the waste water per ton of incinerated waste are almost identical for incinerators with sewage-generating flue gas purification systems. The amounts are on the order of 3 - 4 kg/t of waste. Consequently, an average of some 60% of the HCl in the raw gas is discharged into the waste water. Due to the high solubility of chlorides in water, a similar waste input will produce comparable loads in the waste water irrespective of the water volumes used.

As is shown in table 2, also the sulfur loads in the raw gas are on the same order of magnitude in the MSWI studied. In the figures the mass flows vary between 1.5 and 3.8 kg/t of waste.

Unlike the volumes of chlorine, the sulfur loads to be discharged show major differences for MSWI A

and MSWI B in figure 10. In MSWI B, the mean sulfur load of 0.56 kg/t of waste is almost four times higher than the corresponding load in the waste water of MSWI A. This is a result of the much larger volumes of water used in MSWI A which dissolve a considerably larger amount of gypsum. Moreover, it must be taken into account that high chloride concentrations in the waste water further increase the solubility of gypsum. In MSWI B, almost 60% of the sulfur in the gas are discharged versus less than 15% in MSWI A.

A comparison made for mercury shows the mercury load in the raw gas is on the same order of magnitude in all plants. The mean loads, as shown in table 3, are in the range of 1 - 2 g of Hg/t of waste.

While MSWI A feeds less than 0.001 g of Hg per ton of waste in the waste water, MSWI B discharges approximate 0.5 g/t of waste into the sewage treatment plant. It should be borne in mind that the balance for MSWI B does not include the sewage treatment plant: the partial return of the mercury thus is not taken into account.

Waste incineration plants with wet flue gas purification need auxiliary chemicals to remove the pollutants. This applies in particular to neutralizing agents and precipitation agents for removing heavy metals. MSWI A and MSWI B use calcium-bearing neutralizing agents and NaOH to neutralize the acid pollutants HCl and SO_2, while MSWI C employs a dolomite-bearing neutralizing agent. The requirement for NaOH amounts to approximate 3.7 kg/t in MSWI A and 3.8 kg/t in MSWI B respectively. Both plants show good agreement in consumption.

No exact comparison of the inputs of calcium- and magnesium-bearing agents, respectively, is possible because, on the one hand, different materials are used and, on the other hand, the chemical composition and the water content of the neutralizing agents are not known in most cases. More information can be taken from the stoichiometric ratio of equivalent neutralizing agent to equivalent "acid pollutants," by which the excess of neutralizing agent in the plants can be calculated.

A stoichiometric ratio above 1 in figure 13, is indicating an excess of neutralizing agents. In MSWI A and MSWI C, an excess of neutralizing agents is employed. In MSWI A, the excess is approximately 50%. In MSWI C, on the other hand, the quantities of neutralizing agent used are somewhat on the low side. This is probably due to uncertainties in the model calculation estimating the scrubbing water volume flows, and in the precise composition of the neutralizing agents.

Also precipitating agents for heavy metals are used in the three MSWI in addition to neutralizing chemicals. Comparison is possible only between MSWI A and MSWI C, as both plants use a 15% solution of trimercaptotriazine (*TMT-15®*). This is of interest because the two plants include one sewage-generating and one sewage-free plant. While the consumption data per ton of waste of 0.12 and 0.16 kg of *TMT-15®* do not differ greatly, relating the consumption data to the mercury load in the raw gas from which filter ash has been removed produces the result shown in figure 14.

The *TMT-15®* consumption normalized for the mercury content is clearly higher in MSWI C than in MSWI A. Although heavy metals other than mercury have an impact on consumption, the *TMT-15®* consumption is likely to be based in a first approximation on the mercury load. The higher *TMT-15®* consumption in MSWI C seems to be a consequence of, among other factors, the stability of the mercury-TMT complex in the temperature range around 230°C. This can be seen from operating experience. In the light of elevated Hg concentrations in the raw gas downstream of the spray dryer it may be assumed that evaporation of the waste water from the scrubber in the spray dryer (non-sewage generating operation) partly destroys the mercury-TMT compound.

Conclusions

The studies have shown that from MSWI currently in operation relatively few data about flue gas purification system were available which could be used to describe the material flows in the respective flue gas cleaning devices and to calculate mass balances. The gaps must be filled by plausibility assumptions and model calculations.

Moreover, the balances performed of three different MSWI has shown that the material flows in flue gas purification systems are less dependent on whether the flue gas purification system is operated in the sewage-generating or sewage-free mode. They are decisively influenced by the management of the flue gas purification system (such as the amounts of absorption agent used).

It can also be seen from the studies conducted that especially the amount of chemicals employed, the sulfate load in the waste water, and the heavy metal loads in the waste water can be influenced by plant management: while the high solubility of chlorides causes a more or less constant chloride load, the gypsum load depends on the volume of effluents. The heavy metal loads are determined not only by the waste water volumes, but are also depending on the efficiency of the heavy metal precipitation.

Plant control is based on such parameters as pH, conductivity, and density of the scrubbing liquids and, in most cases, only on clean gas values. For this reason, plant operation is not always optimally adapted to the conditions prevailing in terms of material flows at that time. Hyperstoichiometric additions of auxiliary chemicals, which cannot be excluded entirely at times, cause increased material flows in the plants, thus adding to the salt loads in the sewage, the volumes of residues to be landfilled and, as a result, also the cost.

Figure 1. Chlorine balance of plant A; values given in kg Cl/t waste.

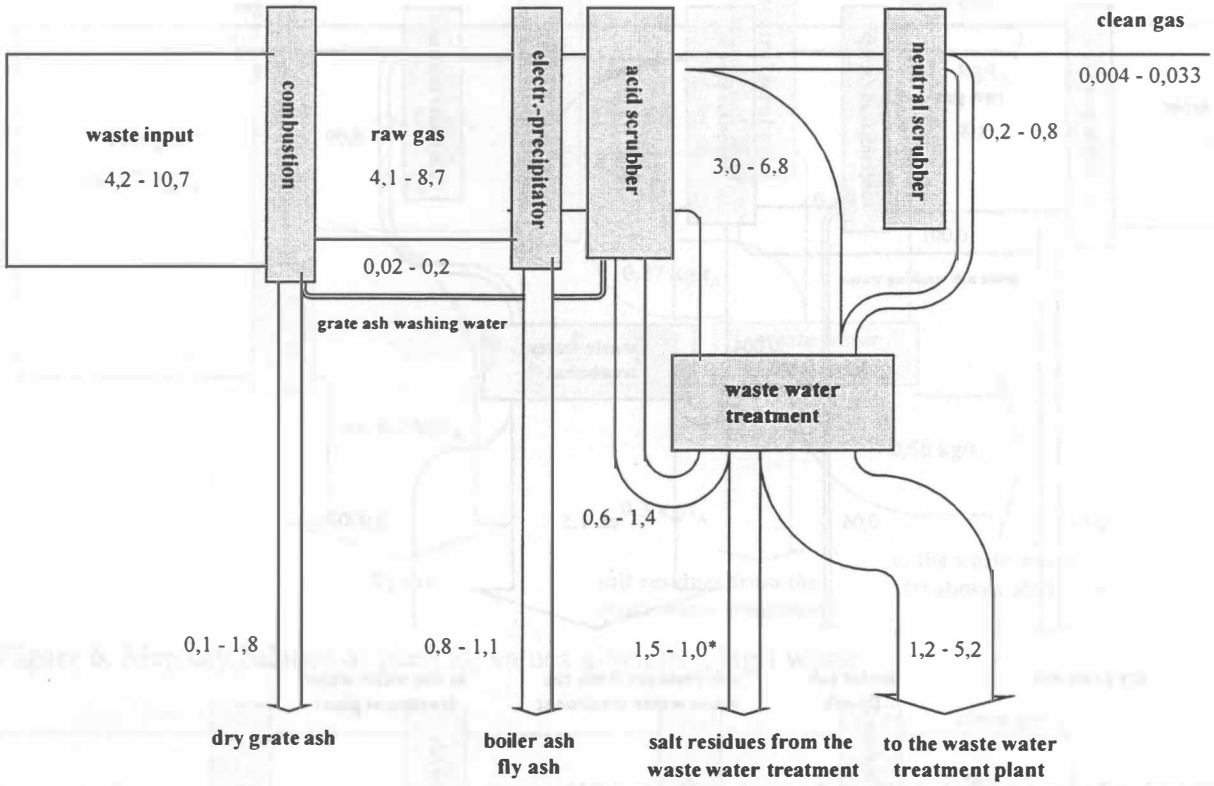

Figure 2. Sulfur balance of plant A; values given in kg S/t waste.

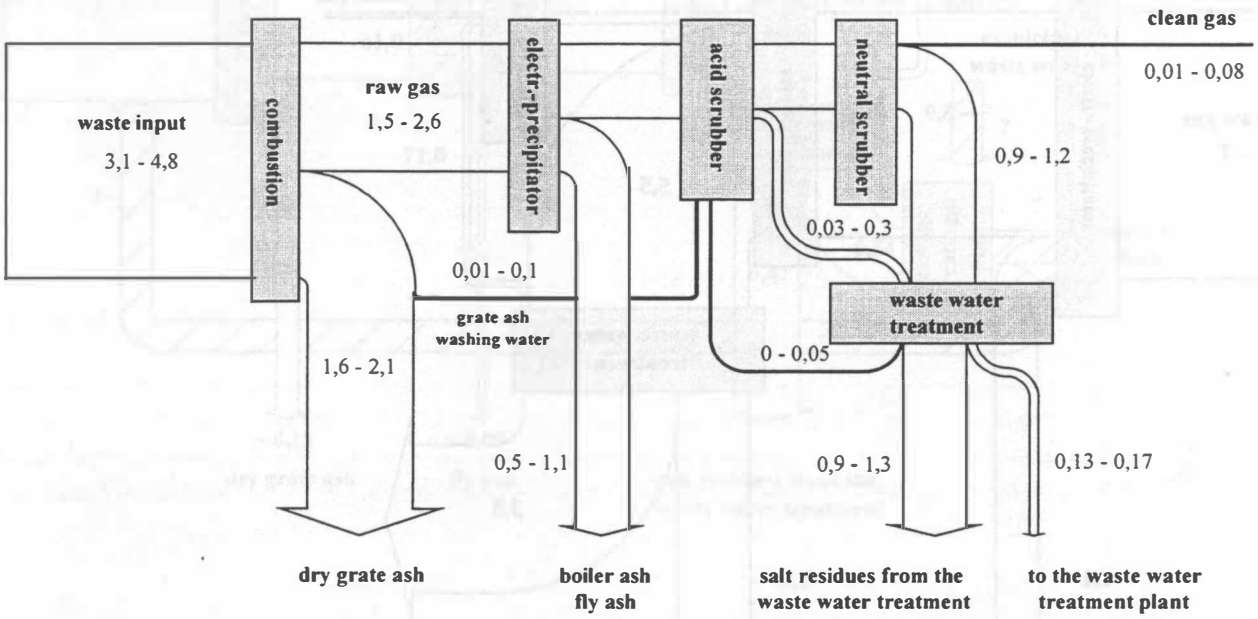

Figure 3. Mercury balance of plant A; values given in g Hg/t waste.

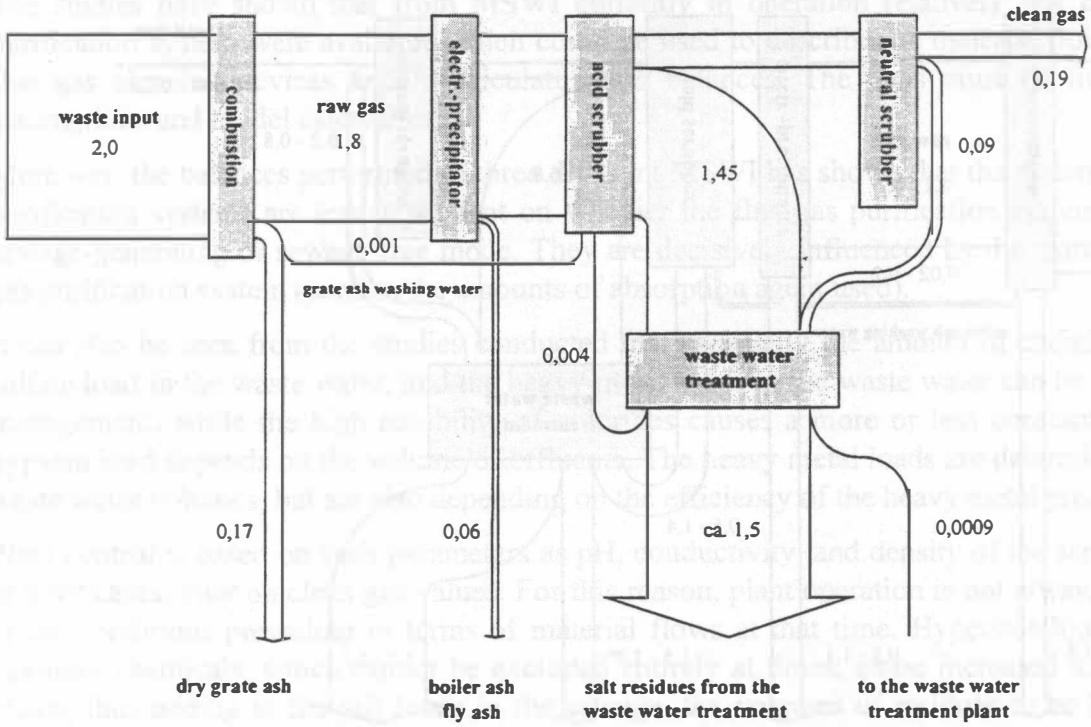

Figure 4. Chlorine balance of plant B; values given in kg Cl/t waste.

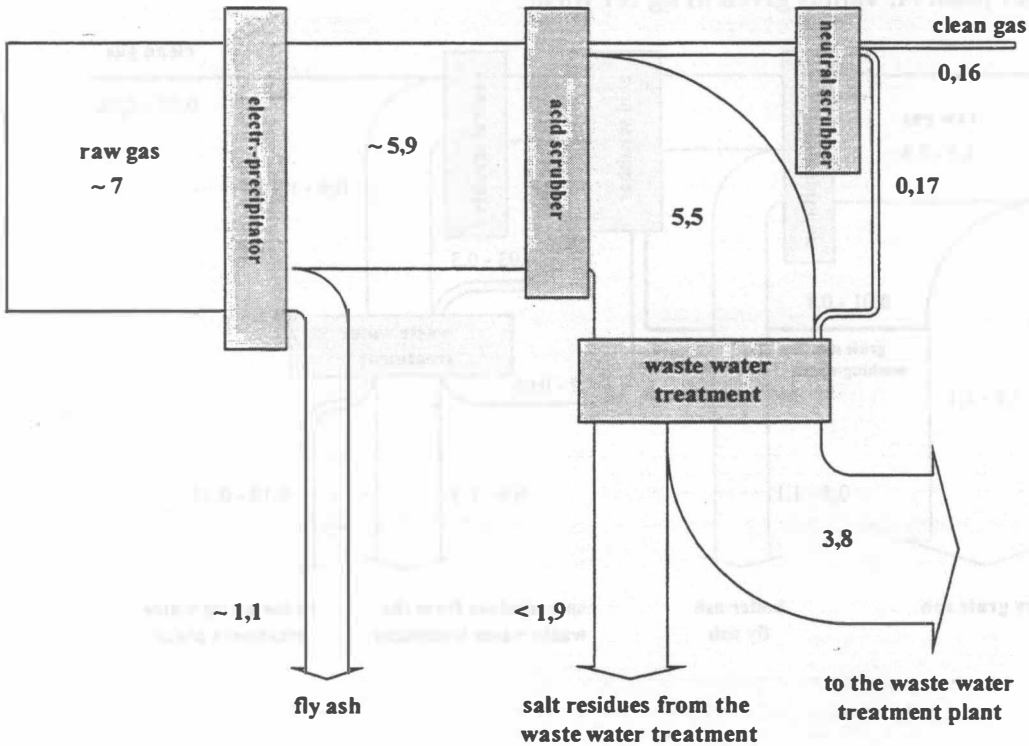

Figure 5. Sulfur balance of plant B; values given in kg S/t waste.

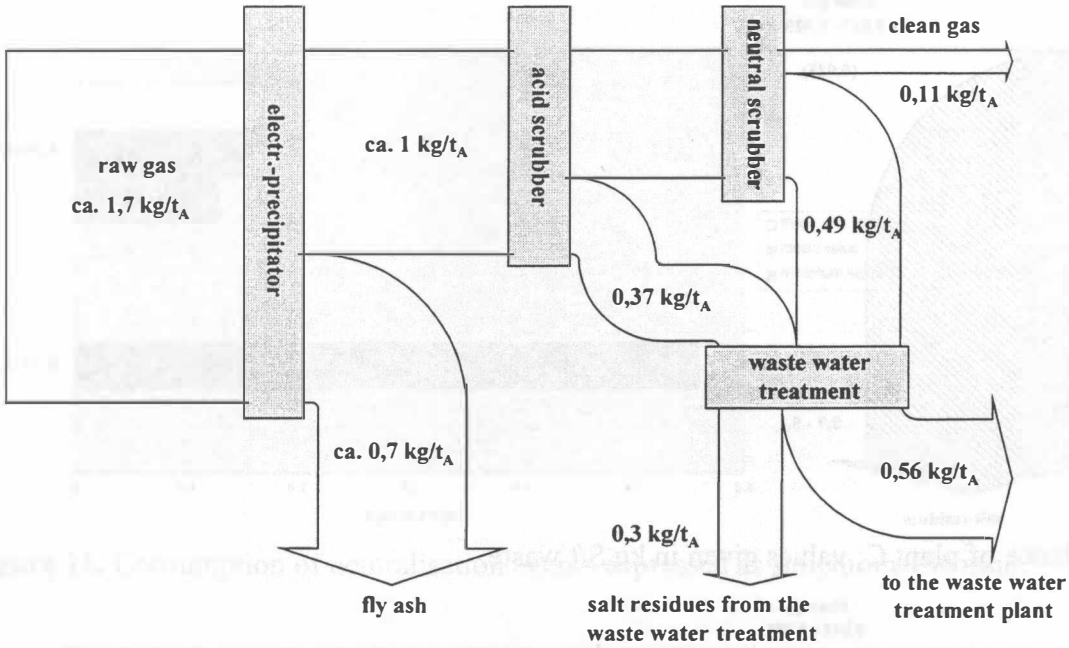

Figure 6. Mercury balance of plant B; values given in g Hg/t waste.

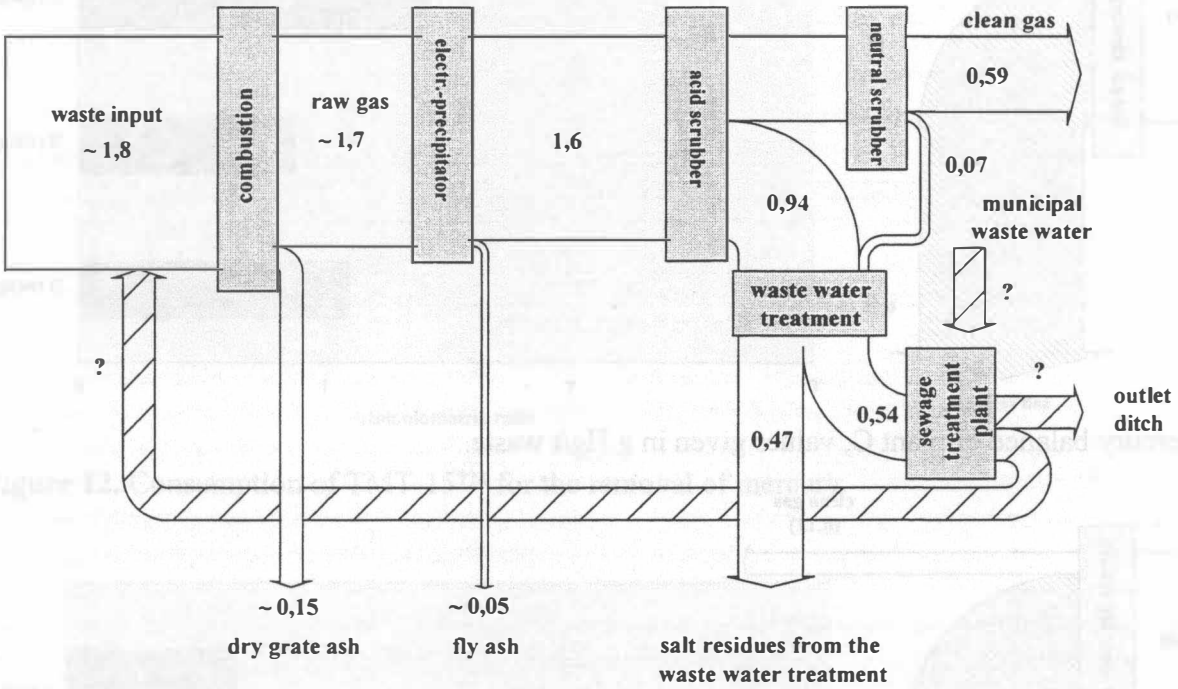

Figure 7. Chlorine balance of plant C; values given in kg Cl/t waste.

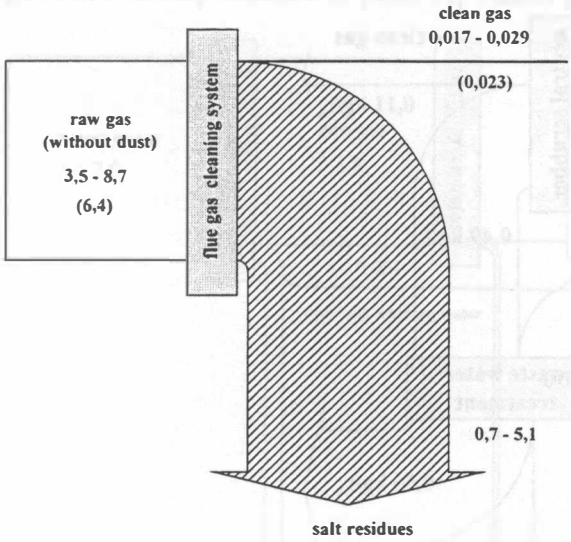

Figure 8. Sulfur balance of plant C; values given in kg S/t waste.

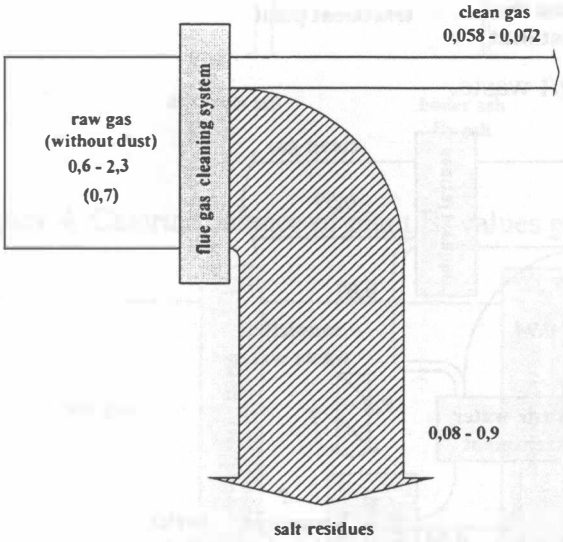

Figure 9. Mercury balance of plant C; values given in g Hg/t waste.

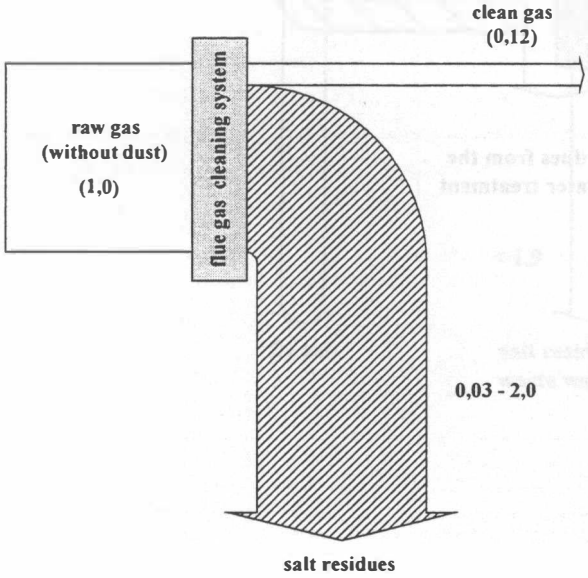

Figure 10. Flow of sulfur in the waste water behind the waste water treatment.

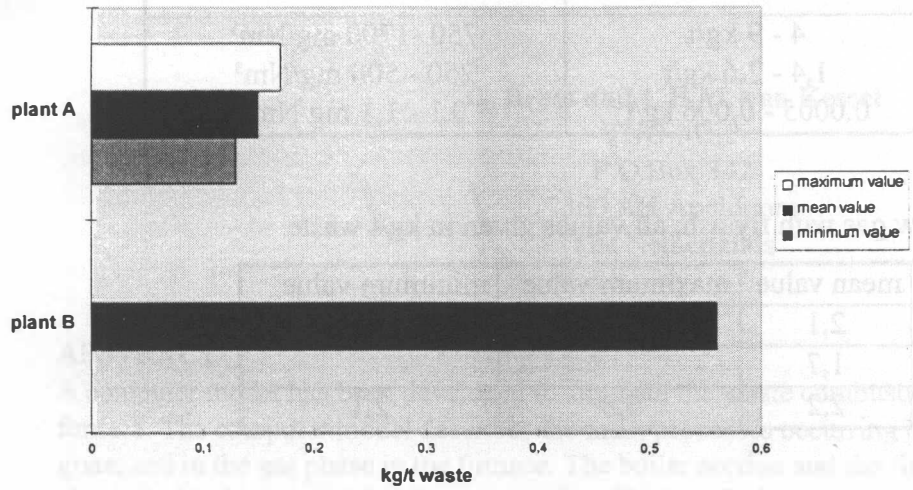

Figure 11. Consumption of neutralisation agents expressed as stoichiometric ratio.

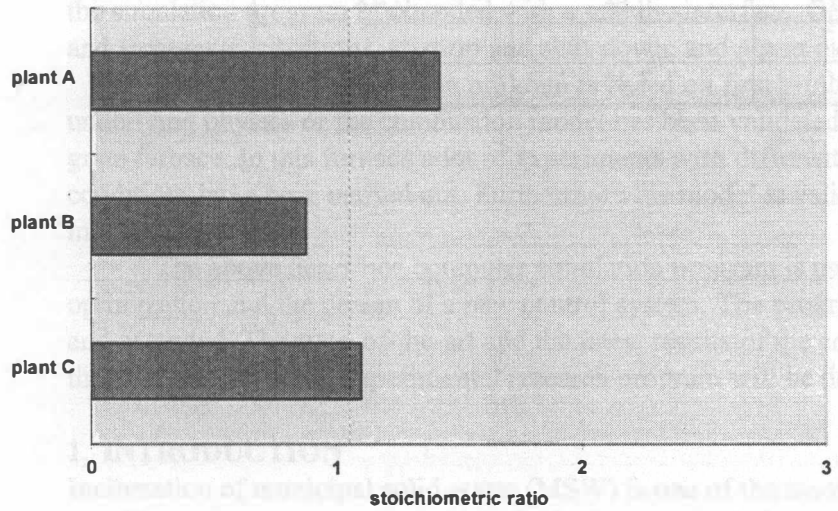

Figure 12. Consumption of TMT-15™ for the removal of mercury.

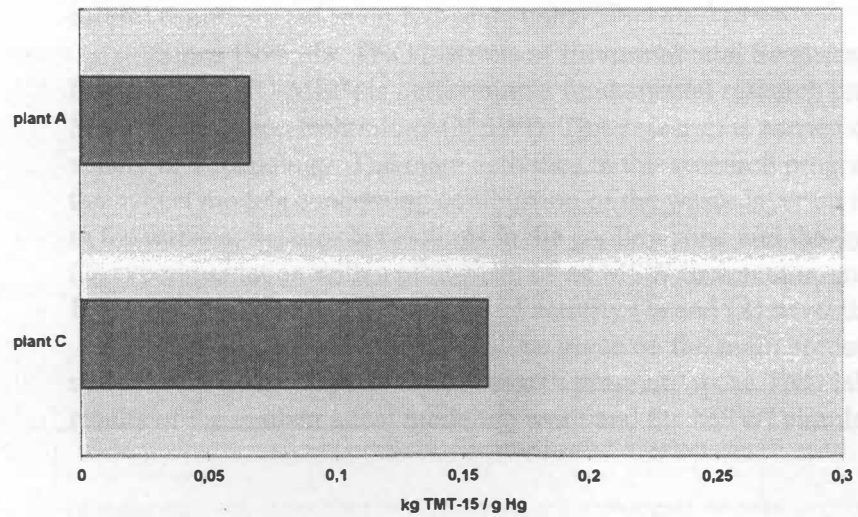

Table 1. Concentrations of pollutants in the raw gas with fly ash.

pollutant	material flow	concentration
chlorine	4 - 9 kg/t	750 -1700 mg/Nm³
sulfur	1,4 - 2,6 kg/t	250 - 500 mg/Nm³
mercury	0,0005 - 0,006 kg/t	0,1 - 1,1 mg/Nm³

Table 2. Flow of sulfur in the raw gas with fly ash; all values given in kg/t waste.

	mean value	maximum value	minimum value
plant A	2,1	2,6	1,5
plant B	1,7		
plant C	2,2	3,8	2,1

Table 3. Flow of mercury in the raw gas with fly ash; all values given in g/t waste.

	mean value	maximum value	minimum value
plant A	1,79	5,54	0,48
plant B	1,7		
plant C	1,0	3,1	0,3