Optimization of additives for flue gas cleaning of sewage sludge incineration

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Abstract

The incineration of sewage sludge has proven to be a reliable solution for the treatment of sewage sludge. Treatment of the flue gas from the incineration of sewage sludge is required to reach acceptable emissions that meet the limits for the EU waste incineration directive 2000/76. In the past years SNB has optimized it´s flue gas cleaning system to achieve low emissions at reasonable costs. This paper gives the results of optimization of the use of limestone in the furnace for capture op sulfur dioxide and of use of mineral adsorbents to capture mercury.

Keywords: sewage sludge, incineration, fluidized bed, desulfurization, sulfur dioxide, high temperature, lime stone, mercury, mineral adsorbent

Introduction

In the last decades there has been a growth in the incineration of sewage sludge. In the past this sludge was mainly used as a fertilizer in agriculture, but contamination of the sludge with heavy metals and organic pollutants, especially persistent organic pollutants (POP's), has raised concern whether the sludge can be used safely as a fertilizer. In The Netherlands the use of sewage sludge in agriculture has not been possible since 1995. In other European countries and according to European directives sewage sludge can be used in agriculture. Nevertheless there is an increasing trend to incinerate the sludge, especially in Northern Europe. The sludge can be incinerated in mono-incineration plants for sewage sludge or co-incinerated in coal fired power plants or cement kilns. Mono-incineration of sewage sludge produces a fly ash that can be used as a filler material in road construction. The fly ash is rich in phosphates and recent research [1, 2] has produced promising propositions to recycle these phosphates thus enabling the conservation of a scarce resource in a safe manner.

Mono-incineration of sewage sludge is mainly performed in bubbling fluidized bed furnaces as these furnaces have proven to reliably incinerate sludges with low heating values (3-4 MJ/kg). Control of the emissions resulting from the incineration of the sludge is important and requires an extensive flue gas cleaning system. The main points of concern are the emissions of nitrogen components (NO_x, N_2O and NH_3), acidic gases (mainly SO_2 as well as HCl and SO_3) and mercury. Due to the very effective incineration in the fluidized bed emissions of unburned hydrocarbons, carbon monoxide and dioxins are hardly a problem. The emission of the nitrogen emissions can be controlled through

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the incineration conditions (temperatures, air-to-fuel ratio) in the furnace. As this article focuses on additive use this is not the subject of this article. More information can be found in [3].

The reduction of the emission of acidic gases can be achieved in several ways. At SNB limestone is dosed to the furnace to capture sulfurdioxide with the fly ash at high temperatures. In this way already 60-70% of all sulfurdioxide is captured in the furnace. This article will describe work that has been done to optimize the use of limestone for this application.

Due to the low chlorine content of sewage sludge the flue gas contains a relative high amount of metallic mercury. The metallic mercury is more difficult to capture than ionic mercury and at SNB the mercury is captured by dosing a mixture of adsorbent and hydrated lime in a bag house filter as a last step in the flue gas cleaning.

Process description

SNB operates four incineration lines for the treatment of sewage sludge from municipal waste water treatment plants. On an annual basis 420 000 tons of wet sludge (100 000 t/y as dry matter) is incinerated in the installation. This amount represents 28% of the total sludge production in The Netherlands and is produced in more than 70 waste water treatment plants representing a total capacity of approximately 8 million people equivalents. The installation is one of the largest sludge incinerators in Europe.

After pre-drying the sludge to a dry matter content of 35-40% the sewage sludge is combusted in four identical bubbling fluidized bed furnaces. The bed temperature is normally maintained between 800 and 850°C. The freeboard temperature is maintained within a range of 890 to 940°C.

Limestone is added to the furnace to capture SO_2 that is formed in the furnace. It reacts via calcium oxide to anhydrite ($CaSO_4$) that leaves the furnace together with the fly ash. Part of the calcium oxide and anhydrite remain in the furnace and make up a large part of the bed material. A diluted ammonia solution is injected at the top of the furnace to control the emission of NO_x. Characteristic for the incineration of sewage sludge is the large degree of volatiles giving a large temperature difference between the bed and the freeboard. The dry matter in the sludge has an ash content of 32% giving a large fly ash production. The ash has a high content of calcium, iron and aluminum. These will be present as oxides or phosphates. The fly ash is collected in the steam boiler and in an electrostatic precipitator (ESP). The steam boiler recovers the heat from the incineration process as steam (10 bar, 180 °C) and this steam is used to pre-dry the sludge. Circa 25% of the steam production is a surplus and is used to generate electricity in a steam engine.

After the ESP the flue gas is cooled in a gas-gas heat exchanger from 200 to 150°C with cold flue gas from the scrubber. From the heat exchanger the flue gas is led to a scrubber consisting of an acidic scrubbing (pH =1-2) and an alkaline scrubber (pH =7,2). The acidic scrubber is a spray tower with no packing and also cools the flue gas to a

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temperature of 75°C. The alkaline scrubber is a packed bed scrubber and the pH is maintained by dosing caustic. The bleeds from both the acid and alkaline scrubber are
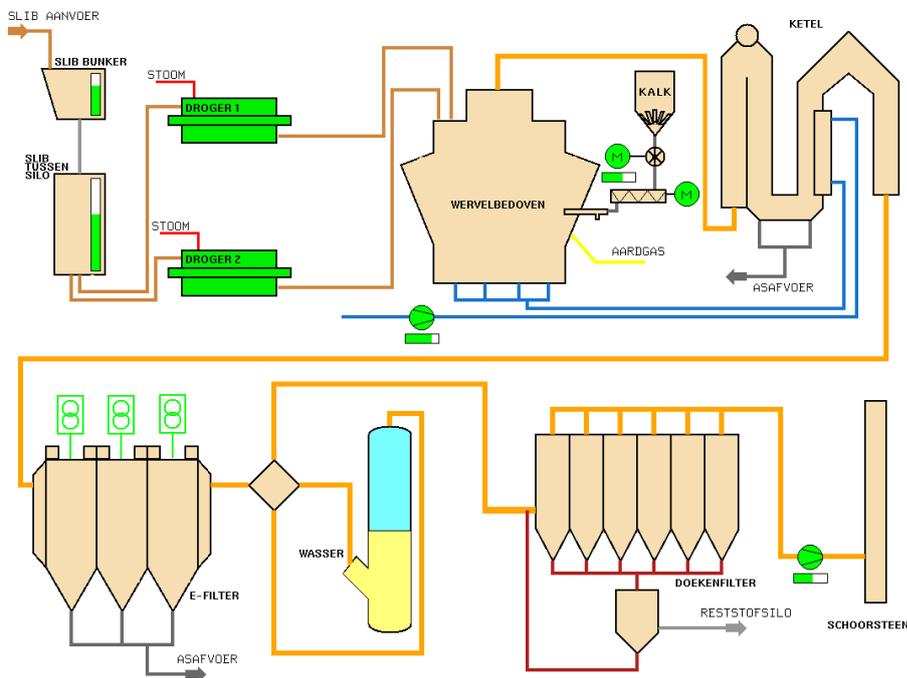

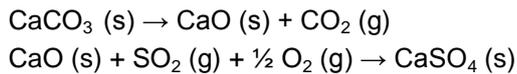Figure 1: process scheme SNB

combined and evaporated in an evaporation plant to produce a salt.

The wet gasses from the scrubber are reheated to 110°C in the already mentioned gas-gas heat exchanger with the hot flue gas coming from the ESP. In the final step of the flue gas cleaning an adsorbent is added to remove any remaining metallic mercury from the flue gas. The adsorbens is removed in a baghouse filter and then partly recycled. The adsorbent also polishes any remaining SO_2 that is still in the flue gas. After this final stage the flue gas can then be emitted through the chimney. Circa 50% of the flue gas is not emitted but transported by a 700 meter pipeline to a neighboring company. This company uses the CO_2 in the flue gas to produce precipitated calciumcarbonate (PCC). This PCC is sold has an high grade whitening agent for the paper industry.

High temperature lime stone addition for desulfurization

The sewage sludge that is incinerated in the furnaces of SNB has a sulfur content of approximately 1%. Without any flue gas cleaning the SO_2-concentration in the flue gas would be circa 3000 mg/Nm^3 based on dry gas. Through addition of limestone to the furnace the SO_2-concentration in the flue gas is maintained at 900 mg/Nm^3. For this the SO_2-concentration is measured after the ESP and this measurement controls the feeding rate of the lime dosing screw.

The reactions taking place in the furnace can be described as follows:

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$$CaCO_3 \text{ (s)} \rightarrow CaO \text{ (s)} + CO_2 \text{ (g)}$$
$$CaO \text{ (s)} + SO_2 \text{ (g)} + \tfrac{1}{2} O_2 \text{ (g)} \rightarrow CaSO_4 \text{ (s)}$$

First the limestone is decarbonized to calciumoxide. The calciumoxide reacts with sulfurdioxide and oxygen to form calciumsulphate. The calciumsulphate is removed from the flue gas together with the fly ash. Part of the formed calciumsulphate accumulates in the bedmaterial if the particle size is too large. The literature describes several parameters that can have an influence on the effectiveness on the high temperature removal of SO_2.

Theoretical backgrounds

Most literature focuses on the desulfurization of flue gas in coal fired fluidized beds rather than with sewage sludge, which makes the findings difficult to transfer to this specific situation. Lyngfelt and Leckner have done extensive research on the capture of SO_2 in fluidized bed furnaces [4,5 & 6]. Their research shows that reducing conditions in the furnace can lead to a significant release of already captured SO_2. This is of importance because the limestone is added to the fluidized bed before the secondary air is entered to the free board. Too low air-to-fuel ratio's can therefore lead to reducing conditions and release of SO_2 as a consequence. Research by Lyngfelt and Leckner showed significant releases of SO_2 during changes between oxidizing and reducing conditions [4].

Their research also shows that temperature is a parameter with an even larger influence [5]. Between 800-850°C reasonable removal rates and additive-efficiency can be achieved. At temperatures higher than 900°C the efficiency and effectiveness is very low. Another important factor for the removal efficiency is the Ca/S ratio in the furnace [4]. Theoretically 1 mol of $CaCO_3$ is required to remove one mol of SO_2, but at such a ratio the SO_2 removal efficiency will normally be not higher than 20-40%, depending on the temperature. To achieve reasonable removal rates of 60-80% the Ca/S molar ratio should be between 2-3.

Particle size of the limestone is another important factor. The smaller the particle size the higher the surface area for the reaction to calciumsulphate, but too small particles will have little residence time in the furnace and will therefore hardly react as is shown by Bragança et al. [7]. Very large particles will have a long residence time but will not mix efficiently in the fluidized bed. The ideal particle size will therefore be a size that is just not blown away from the fluidized bed. The sulphation reaction will cover the limestone particles with a skin of calciumsulphate which in addition has a larger volume than the orginal calciumcarbonate. According to research by Lyngfelt and Leckner [6] this skin can be very resistant to abrasion.

Finally reactivity of the limestone is of importance. Generally the reactivity is associated with the age and consequently the hardness of the mined limestone. Limestone from the Cretaceous period (chalk) is relatively young speaking in geological terms and has a higher natural reactivity than limestone from the Devonian period [8]. The reactivity of this Devonian limestone can be increased by burning the limestone to calciumoxide and a recarbonization to calciumcarbonate. The reactivity of the limestone is also influenced by

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the content of magnesiumcarbonate. Bragança et al. report that limestone with a small content of magnesium (ca. 2%) is more reactive, but generally higher magnesiumcarbonate concentrations will lower the reactivity of the limestone [8]. Magnesiumcarbonate will not react very well with SO_2 as the optimum temperature for SO_2 capture with magnesiumcarbonate is around 700°C, which is too low for incinerators [9].

Experiments

On the furnaces of SNB different experiments have been performed to test the effect of temperature and different limestone additives on the removal of SO_2 in the furnace. The limestone additives where added to the furnace through a special temporary silo. This silo fed the limestone directly to the limestone dosing silo of one of the furnaces. Each additive was tested for at least 24 hours to see direct effects on the removal of SO_2. This time is enough for a first selection of potentially interesting additives, but not enough for a full evaluation, because it will take several days before the content of the fluidized bed is replaced.

The table below shows the tested additives. For each additive also the neutralization value and the Sauerbeck reactivity (comparable to VDLUFA II, 6.4) for sulfuric acid is reported. Sorbacal A was the limestone that SNB had been using in the last years.
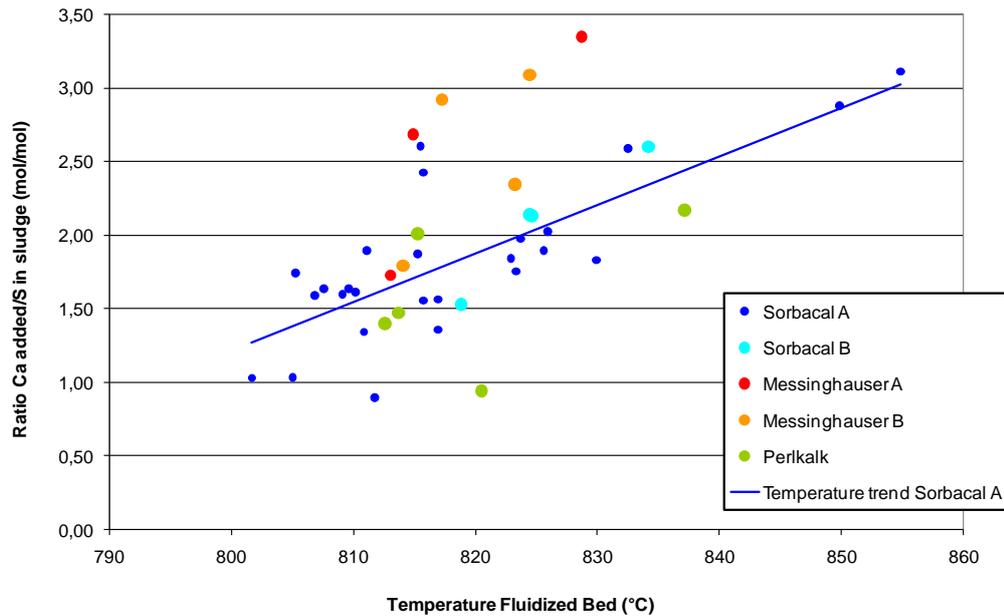
Table 1: characterization of the tested limestones

Limestone additive	Supplier	Characterization	Particle size (mm)	Neutralization value	Sauerbeck reactivity	
					2 min.	10 min.
Sorbacal A (reference)	Rheinkalk	Activated limestone	0,7 - 1,6	57%	39	72
Sorbacal B	Rheinkalk		1 – 2			
Messinghauser A	Rheinkalk	Devonian, hard	0,6 – 0,9	56%	10	33
Messinghauser B	Rheinkalk	limestone	0,9 – 1,2			
Perlkalk	Damman	Chalk, soft limestone	0,5 – 2	50%	25	64

Based on the results of these screening tests one alternative additive (Perlkalk) was tested for a full scale evaluation. For this test the new additive was used on all four incineration lines during a period of 5 days. The objective of this test was to confirm the results of the screening test.

Results

Figure 2 shows the results of tests to determine the influence of the temperature and type of limestone on the efficiency of the limestone addition. First of all the figure shows the significant effect of the temperature of the fluidized bed on the limestone consumption as shown by the data for Sorbacal A. A temperature increase of 20°C will increase the consumption of limestone with more than 30%. The best correlation was found with the temperature in the fluidized bed. The correlation with the free board temperature was significantly less and shows that the relevant reactions are more likely to take place in the fluidized bed than in the freeboard. This is of importance because one strategy for the reduction of the emission of N_2O is to increase the freeboard temperature. By lowering

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Figure 2: effect of temperature and type of limestone

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the air-to-fuel ratio the free board temperature can be increased, while maintaining low fluid bed temperatures and thus an acceptable limestone consumption [3].

The figure also shows that the hard, Devonian Messinghauser limestone is clearly less reactive than the activated limestone that is normally used. Even if the lower cost price of this limestone is taken into account it is not a better alternative due to the higher consumption and the higher fly ash production. A broader range for the particle size of Sorbacal showed no significant differences within the accuracy of the measurements. This alternative is slightly cheaper than Sorbacal A and therefore it was decided to completely change-over to this alternative after the tests. The long term results confirmed these test results. The tests also showed that the chalk alternative (Perlkalk) could possibly compete in efficiency with Sorbacal A. Because chalk has a natural high reactivity it is significantly cheaper than Sorbacal A and therefore an interesting alternative. Another advantage is that the production of chalk is less energy-consuming than the production of Sorbacal A, where the limestone is first roasted to produce CaO. The results also show that the reactivity in sulfuric acid (Sauerbeck analysis) gives a good first impression of the behavior in the furnace although the reaction conditions are significantly different.

Based on the results of this first screening it was decided to perform a larger test where the chalk alternative was tested on all 4 incineration lines during 5 days. These tests showed a consumption of limestone that is comparable to the results with Sorbacal A. The tests did show a tendency to a higher production of fluidized bed ashes, which is unwanted because of the larger disposal costs for these ashes compared to fly ash. In addition the analyses of the fly ash showed a lower sulfur content and that would suggest the accumulation of sulfur in the fluidized bed. Therefore a final test with was to be done for the final evaluation. With this test the original limestone was fully replaced with the chalk alternative during a period of 4 weeks. Table 2 shows the results of this test. During

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the test the limestone consumption and the fly ash production were more or less similar to the results with the normally used limestone, Sorbacal A. A significant difference can be seen in the bed ash production for the chalk alternative. This production rate was too high to cope with for the normal removal systems. Based on these experiences it was decided to only replace at maximum half of the limestone by the chalk alternative. This has been realized from March 2009 to this date. The results until now confirm the results above. This replacement has generated an annual benefit of more than 100.000 euro.

Description	Limestone usage (kg/ton wet sludge)	Fly ash production (kg/ton wet sludge)	Bed ash production (kg/ton wet sludge)
4 weeks before test (Sorbacal A)	20	92	3,7
4 weeks of test (chalk, Perlkalk)	15	79	7,3
4 weeks after the tests (Sorbacal A)	16	83	4,2

Table 2: results of large scale test with chalk alternative.

Mineral adsorbent for Hg-removal from flue gas

As described in the process description the last step in the flue gas treatment of SNB is the addition of a mixture of cokes and limestone. The adsorbent is added to the flue gas as a mixture of 10% cokes (hertofencokes) and 90% hydrated lime. The hydrated lime is added to the mixture as a safety measure to prevent dust explosions as it makes the total mixture non-explosive. In addition the lime has the benefit of polishing any remaining SO_2 and HCl from the flue gas. This also prevents corrosion of the baghouse filter that is used to remove the adsorbent mixture from the flue gas.

Figure 3 shows the process diagram of the adsorbent installation at SNB. The figure shows that the recovered adsorbent from the bagfilter is reused and that only a percentage (normally 20%) is continuously replaced by a fresh adsorbent mixture. Since 2005 the spent adsorbent mixture is partly (for circa 50%) re-incinerated in the fluidized beds to reduce the amount of waste material.

Normally 125 kg/h of adsorbent-mixture is added to the flue gas giving a dosage of circa 6 g/Nm^3 (dry gas @ 11%O_2). Continuously 25 kg/h of fresh adsorbent-mixture is added to the system. The bagfilter removes 30-40% of the total mercury that is released from the sewage sludge. The mercury concentration in the inlet to the adsorbent stage can vary from values as low as 20 µg/Nm^3 (dry gas, @11% O_2) to 150 µg/Nm^3. The emission limit for mercury is 50 µg/Nm^3 (dry gas, @11% O_2) and normally SNB achieves an emission concentration of 0-5 µg/Nm^3 (dry gas, @11% O_2). Dioxin and furan concentrations to the bagfilter are generally very low due to the good incineration conditions in the fluidized bed furnace. The removal efficiency of the adsorbent for dioxins and furans is therefore not very important for the situation at SNB.

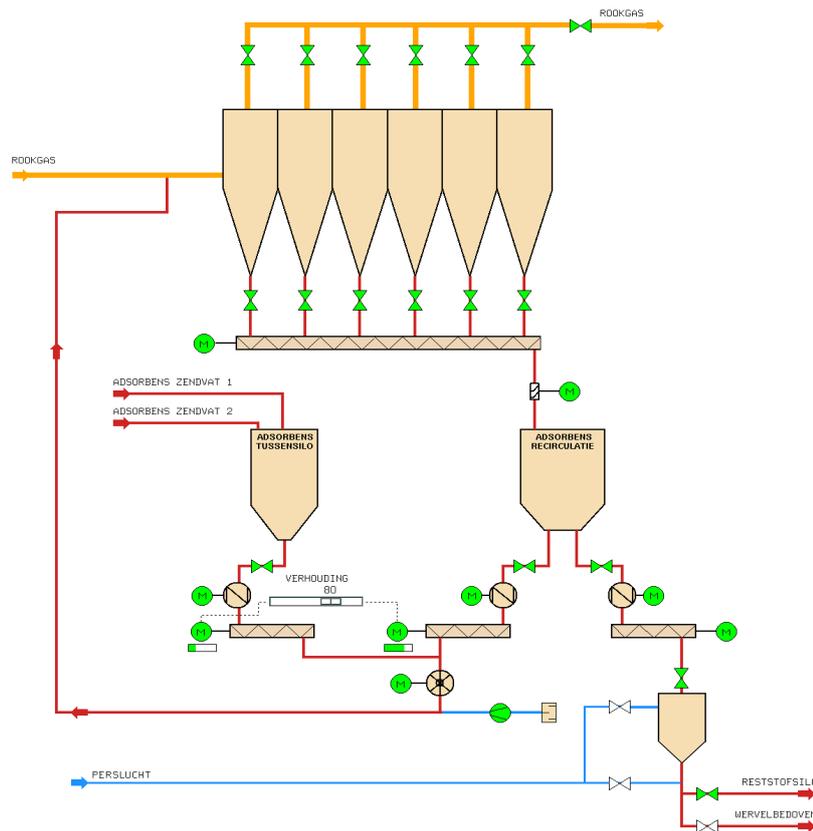Referent: Leon Korving, SNB | Vortrag: Optimierung des Einsatz von Sorbentien bei der Klarschlammverbrennung

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Figure 3: proces diagram adsorbens installation

Safety

The cokes in the adsorbent mixture is combustible and therefore several safety measures are incorporated in the design of the system:

1. Mixture of the cokes with 90% hydrated lime to achieve a mixture that is non-explosive and less combustible.
2. Temperature measurement in all the hoppers of the bagfilter and in the silo for spent adsorbent to detect any unexpected rise in temperature.
3. Measurement of the CO concentration before and after the bagfilter to detect any formation of CO in the bagfilter.
4. All equipment can be fed with CO_2-gas to extinguish hot spots or onset of fires.

Experiences at SNB have shown that these safety measures are required and hot spots can form in certain situations. Especially in the spent adsorbens silo there have been several occasions of the formation of severe hotspots.

Mineral adsorbent

As SNB prefers an inherent safe process, SNB was very interested to test the fully mineral adsorbent Minsorb®-Me that is offered by the Lhoist-company. The mineral adsorbent is based on a clay like material that should have a very similar adsorption capacity as coke for mercury and dioxins. Additional advantages would be that it would

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enable to lower the content of lime in the mixture, thus achieving a cost benefit as well as a lower waste production.

First tests were performed with a temporary installation that fed the Minsorb to one of the incineration lines in January and February 2011. The objective of these tests was to prove that a similar mercury removal could be obtained and that no operational problems would occur. During these tests mercury was continuously measured in the exit of the baghouse filter by Lhoist. In addition secondary measurements were carried out by an external measuring company at the end of the tests.

Based on the results of the tests it was decided to fully changeover to the Minsorb alternative on all incineration lines in April 2011. To verify the effect on the mercury emissions the changeover was done more than a week before the regular emission measurements. Because the pilot experiments showed a slight increase in the mercury emission a 20% Minsorb mixture was used in the full scale test. This would enable to increase the dosage of adsorbent to lower the emission. To check the influence of the recirculation ratio two lines (line 1 and 2) where operated at a lower refreshment ratio than lines 3 and 4. Based on the results of these measurements it was decided to operate all lines at the lower refreshment ratio (10% fresh/90% old). A half year later the next regular mercury measurements were performed in September with these conditions.

Results

Table 3 summarizes the results of the mercury measurements that have been performed during the changeover to the mineral adsorbent.

During tests C (pilot test on line 2) and D (full scale test) also external mercury measurements were performed upstream of the bagfilter. The measurement during the pilot test (C) showed a surprisingly low mercury concentration of 7-10 µg/Nm^3 (dry gas, @11% O_2). As in the past never such low mercury concentrations had been measured, these measurements were considered to be incredible. During the large scale implementation (test conditions D) again upstream measurements were done on line 2. These measurements showed a mercury concentration of 20-23 µg/Nm^3 (dry gas, @11% O_2). This was still a low concentration compared to previous measurements.

Normally dioxins and furans (PCDD/F's) are measured once a year according to Dutch and EU regulations. In 2010 all measurements on all lines were below the detection limit of 0,02 ng/Nm^3 (dry gas @ 11%O_2). During the pilot test (test C) additional measurements were carried out both upstream and downstream of the filter. These measurements showed PCDD/F-concentrations lower than 0,01 ng/Nm^3 (dry gas @ 11%O_2) before and after filter. During the regular emission measurements in September 2011 (test F) all measurements for PCDD/F were lower than 0,02 ng/Nm^3 (dry gas @ 11%O_2).

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Test	Hg emission (µg/Nm^3, dry @ 11% O$_2$)	Remark
A. Average 2010 with 10%HOK, 25 kg/h fresh adsorbent	< 3	24 one-hour external measurements (4 lines, 2*3 measurements/line)
B. Feb. 2011, pilot test line 2 with 10%Minsorb, 25 kg/h fresh adsorbent,	4,5 - 7,1	Results over 6 days of continuous measurement by Lhoist
C. Feb. 2011, pilot test line 2 with 20% Minsorb, 14 kg/h fresh adsorbent	7 – 13 7 – 11	Results over 10 days of continuous measurement by Lhoist 3 one-hour external measurements
D. Apr. 2011, full scale test line 1 & 2, 20% Minsorb, 14 kg/h fresh adsorbent	< 3	6 one-hour external measurements (2 lines, 3 measurements/line)
E. Apr. 2011, full scale test line 3 & 4, 20% Minsorb, 25 kg/h fresh adsorbent.	< 3	6 one-hour external measurements (2 lines, 3 measurements/line)
F. Sep. 2011, full scale test line 1 - 4, 20% Minsorb, 14 kg/h fresh adsorbent.	10 - 28	12 one-hour external measurements (4 lines, 3 measurements/line)

Table 3: results for Hg-measurements during change over to mineral adsorbent.

The experimental results with the mineral adsorbens are not yet conclusive on the removal efficiency for mercury of the mineral adsorbent compared to the cokes based alternative. All measurements show that compliance to the emission limit of 50 µg/Nm^3 (dry gas, @11% O$_2$) appears to be possible. Nevertheless the emission results vary significantly and are on average higher than with the cokes-based adsorbent. As the Dutch authorities consider the results with cokes-based adsorbent "Best Available Technology" they require SNB to achieve the same low mercury emissions with the mineral adsorbent. Therefore more work has to be done to improve the performance of the mineral adsorbent. Because of the safety advantages of the mineral adsorbent SNB will continue to evaluate the feasibility of the use of the mineral adsorbent either by higher dosages or by improvement of the adsorbent together with Lhoist, the supplier of the adsorbent.

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