# Process Optimisation of Quasi-Dry Waste Gas Cleaning with Integrated Dioxin and Furan Adsorption

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## ABSTRACT

The results obtained from operational tests in a refuse incineration plant for municipal and industrial wastes serve to describe activated lignite's efficiency as a mass sorbent for dioxin reduction in the filter-bed process, with use being made of the existing infrastructure. Variations in the dosing rate, on the one hand, and the use of different grain sizes, on the other hand, identify the selection criteria for the determination of the process-engineering optimum for waste gas cleaning, with sorbent injection being integrated into the existing facility of quasi-dry waste gas cleaning. This paper demonstrates that the dioxin emission can be reduced by activated lignite to below 0.1 ng TE/m³ although the available equipment was in no optimum condition.

## INTRODUCTION

Numerous investigations in recent years have considerably enlarged our knowledge of environmental toxicants and their impact on nature and man. This knowledge and the increasing sensitisation of the population to environmental toxicants result in environmental regulations being continuously tightened up and in ordinances prescribing low emission limit values. This includes in particular the emission limit values of organic compounds such as dioxins and furans and those of heavy metals. Since primary measures alone do not permit a sufficient reduction in emissions, additional cleaning facilities have to be installed that often call for high investment and sophisticated technologies. In view of growing competitive and cost pressures, process-engineering solutions are being looked for that do not entail these disadvantages or have them only in some cases. The target is to obtain an efficient and at the same time favourably priced process for emission reduction by means of an integrated waste gas cleaning system.

Cleaning stages for all relevant pollutants are those adsorptive processes that are designed as moving-bed filter with granular sorbents or as entrained-phase filter with pulverized sorbents. Compared with the moving-bed process, the entrained-phase adsorption with activated lignite as a reasonably priced mass sorbent represents a

simple and economical process variant, which--what is more--can be retrofitted in existing plants with low expenditure involved.

This contribution is aimed at demonstrating the possibility of process optimisation for quasi-dry waste gas cleaning with integrated dioxin and furan adsorption by means of activated lignite.

## POLLUTANT RETENTION BY ACTIVATED LIGNITE

Thanks to its porosity and its large inner surface and pore structure involved, activated lignite from Rhenish lignite produced according to the so-called rotary hearth furnace process [1] is suitable as an adsorbent with an efficiency comparable to that of high-quality activated carbons. Table 1 lists the physical and chemical properties of activated lignite essential for its use in waste gas cleaning. With regard to thermal utilisation of the activated lignite employed, e.g. in firing systems, the Table also includes the respective relevant data.

The processes available on the market for adsorptive gas cleaning on activated lignite basis are techniques reliably tested for many years that primarily include the moving-bed, the entrained-phase and filter-bed systems. [2, 3, 4]

The entrained-phase process by means of an electrostatic precipitator or fabric filter studied in the following is marked by the dosing of pulverized activated lignite to the gas flow where the first step of pollutant separation takes place. A downstream dust separator serves to remove the sorbent spent and any existing process dusts. Here, fabric filter systems have the advantage of the adsorptively acting filter bed (see Fig. 1). Further details about the activated lignite-based entrained-phase process are given in [4, 5].
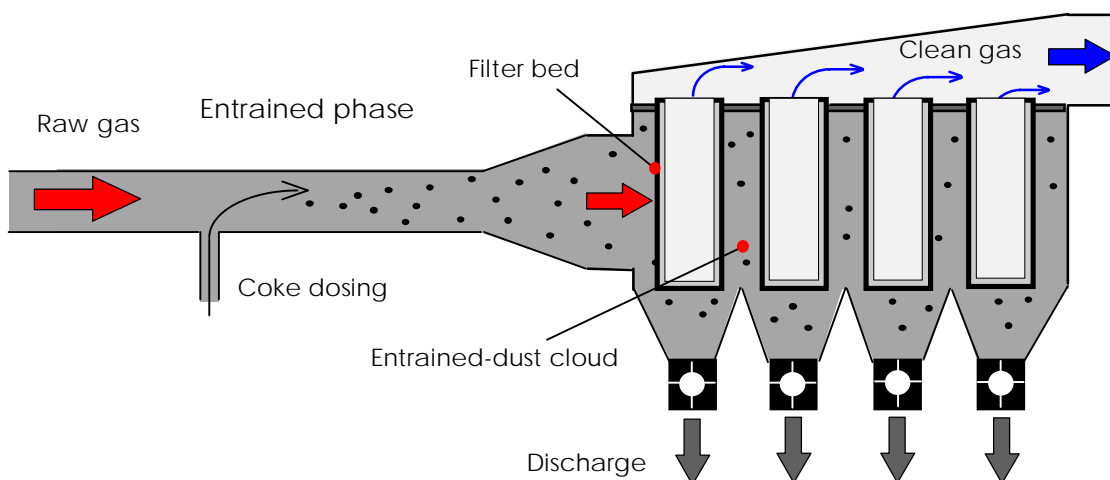
Fig. 1: Filter-bed process

Table 1:    Physical and chemical          parameters of activated lignite

| Physical parameters | Fine activated lignite "HOK grain" | Extra-fine activated lignite "HOK medium" | Pulverized activated lignite*) "HOK pulverized" | Reactivity-enhanced pulverized activated lignite *) "HOK super" |
|---|---|---|---|---|
| Grain size [mm] | 1.25 - 5 | 0.1 - 1.5 | < 0.4 | < 0.2 |
| Bulk density [t/m³] | 0.45 | 0.53 | 0.55 | 0.55 |
| Specific surface | 300 m²/g | 300 m²/g | 300 m²/g | 300 m²/g |
| | | | | |
| Proximate analysis | | Moisture content | 0.5 | % wt |
| | | Ash content | 9 | % wt |
| | | Heating value | 29.9 | MJ/kg |
| | | Volatiles | 3 | % wt |
| | | | | |
| Ultimate analysis | | Carbon | 88 | % wt |
| | | Sulphur | 0.5 | % wt |
| | | | | |
| Ash analysis | | CaO | 41 | % wt |
| | | MgO | 14 | % wt |

*) Activated Lignite qualities used during the tests on PCDD/F separation

## STRUCTURE AND IMPLEMENTATION OF TESTS

The tests were carried out in the existing waste gas line of a refuse incineration plant for municipal and industrial wastes.  Fig. 2 schematically shows the set-up of the incineration line with the sorbent dosing device installed.
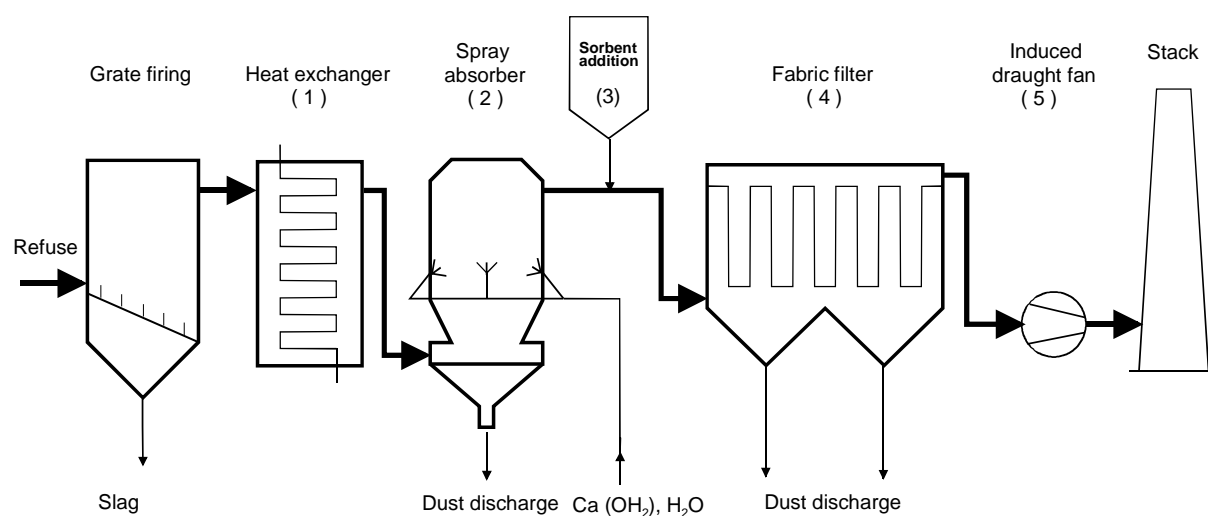
The waste gas line consists of:

- a waste heat unit with heat transfer (1),
- a spray absorption tower with the addition of calcium hydroxide as absorbent for the acid substances contained in the waste gas ($SO_2$, HCl, HF) (2),
- an activated carbon/lignite injection device (3), and
- a downstream fabric filter for dust separation (4).

After dust separation, the clean gas is supplied via an induced draught fan (5) to the stack and then discharged into the atmosphere.

At different dosing rates and with different grain-size qualities, the activated lignite adsorbent is dosed through the existing sorbent injection device to the raw gas flow directly downstream of the spray absorber and upstream of its entry into the fabric filter.  Except for the additive and the dosing adjustment, no modifications were made to the operational sequence nor was the equipment of the plant changed. Thus, a substantial requirement of the investigation into process-integrated adsorption was met by use being made of the existing infrastructure and dispensing with additional investment expenditure.  In order to evaluate the influence of the dosing rate and the grain-size quality on pollutant separation, three tests were carried out with pulverized activated lignite (63 μm) at dosing rates of 50, 150 and

300 mg/m³ and two tests with reactivity-enhanced pulverized activated lignite (median: grain size of 28 µm) at dosing rates of 150 mg/m³ and 300 mg/m³. To obtain steady-state conditions in the case of filter cloth loading, the dosing rate was adjusted to the value specified in the test programme several days before the start of each measurement. As a basis for the comparative tests with activated lignite, the clean gas was first measured during an operation with 50 mg/m³ of activated carbon that is usual for the refuse incineration plant. Later, however, it turned out that the filtering system was not in its proper condition at that time so that this measurement had to be rejected as reference emission.
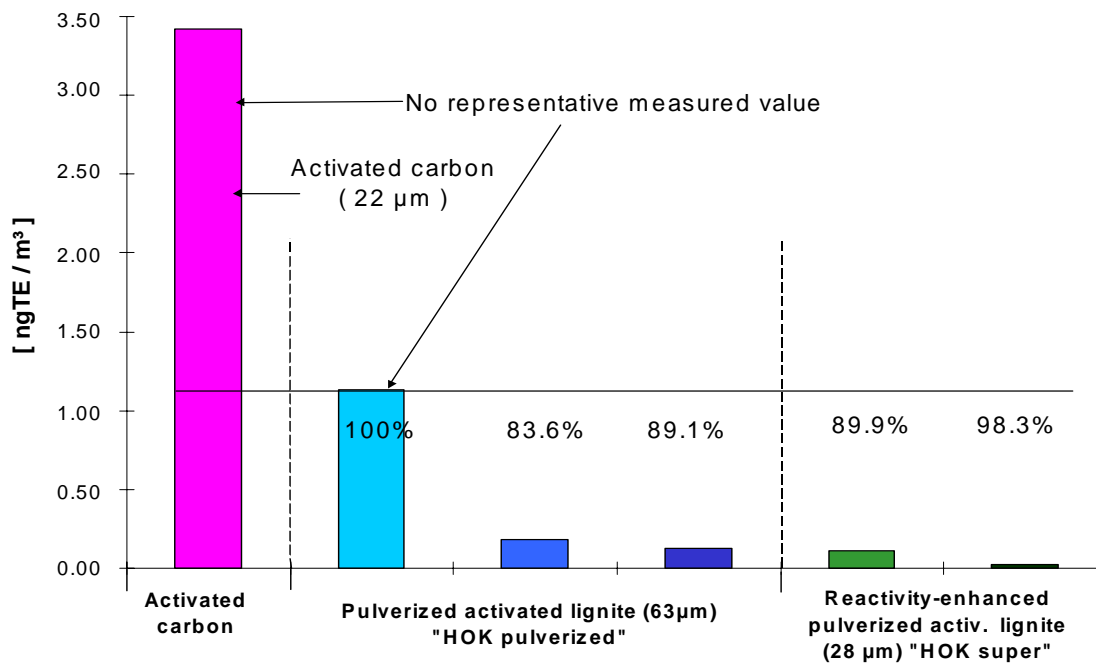
Fig. 2:    Incineration line

Grate firing    Heat exchanger ( 1 )    Spray absorber ( 2 )    Sorbent addition (3)    Fabric filter ( 4 )    Induced draught fan ( 5 )    Stack

Refuse

Slag    Dust discharge    Ca $(OH_2)$, $H_2O$    Dust discharge

| Plant parameters: | |
|---|---|
| Refuse throughput (incinerator): | 4 t/h |
| Waste gas flow rate: | 30,000 m³ (STP), dry/h |
| Waste gas temperature upstream of filter: | 150 °C |
| Process dust content upstream of filter: | 5 g/m³ (STP), dry, on average |

In order to examine and assess the pollutant reduction, samples were taken from the clean gas downstream of the filtering system and analysed for dioxins and furans. In addition, samples were taken from the separated filter dust and analysed for their carbon contents. Besides the additive concentration in the filter dust mixture, this was to check the activated lignite sorbent's equidistribution in the filter.

# RESULTS AND DISCUSSION

Fig. 3 shows the results from the clean gas measurements in respect of dioxins and furans that were carried out during the tests. They are obtained as toxicity equivalents (TE according to NATO/CCMS). Moreover, further data on the test conditions are provided.

Fig. 3: Comparison of dioxin separation (11 % vol $O_2$ reference) for different adsorbents and dosing rates



| | Activated carbon | Pulverized activated lignite (63µm) "HOK pulverized" | | | Reactivity-enhanced pulverized activ. lignite (28 µm) "HOK super" | |
|---|---|---|---|---|---|---|
| Dosing rate [mg/m³ STP, dry] | 50 | 50 | 150 | 300 | 150 | 300 |
| Carbon content, filter dust [% wt] | 1.8 | 1.9 | 3.5 | 6.1 | 2.9 | 5.3 |
| TE/NATO [ng/m3] | 3.42 | 1.13 | 0.18 | 0.12 | 0.11 | 0.02 |
| $O_2$ content [% vol] | 14.6 | 14.3 | 14.7 | 14.2 | 14.6 | 14.9 |
| CO content [mg/m³] | 4 | 1 | 8 | 8 | 7 | 28 |
| Waste gas temp. [°C] | 144 | 146 | 142 | 143 | 143 | 143 |

If we assume that the PCDD/F raw gas conditions were approximately the same during the measurements, the comparison of the PCDD/F concentrations obtained in the clean gas downstream of the fabric filter clearly reveals the influence of the dosing rate and the grain-size quality of activated lignite on the feasible separation efficiency. With the addition of reactivity-enhanced pulverized activated lignite and at a dosing rate of 300 mg/m³ it was possible to attain a PCDD/F value of 0.02 ng TE/m³ in the clean gas which markedly falls below the limit concentration of 0.1 ng TE/m³ prescribed for refuse incineration plants. Striking phenomena here are

the PCDD/F concentrations measured in the clean gas when 50 mg/m³ of activated carbon and 50 mg/m³ of pulverized activated lignite have been added. Compared to the values recorded at higher dosing rates, more than 1 ng TE/m³ is remarkably high in this case. The reason for this is the particle-bound dioxins and furans emitted during these measurements due to the frequent bypass operation; these pollutants are recorded in the case of PCDD/F determination in the clean gas as well.

In the course of the investigation it became obvious that the plant had a considerable optimisation potential for dioxin and furan reduction so that the test series were accompanied by plant optimisation for process-integrated adsorption. For this reason, the individual results can be compared only with certain restrictions and as such are to be regarded as links of an optimisation chain. The influences on PCDD/F reduction observed in the application studied are shown and described in greater detail in the following.

## Influence of Dust Emission on PCDD/F Emission

In order to assess the influence of dust emission on the PCDD/F content in the clean gas, the following Table 2 lists the dust contents measured in the clean gas downstream of the fabric filter. During PCDD/F sampling the measurements were taken with the bypass closed. In addition, further data on filter operation are given.

Table 2: Dust contents during the sampling period of the PCDD/F measurements

| Test series | 1 | 2 | 3 | 4 | 5 | 6 |
|---|---|---|---|---|---|---|
| Adsorbent | AC[1] | AL[2] | AL[2] | AL[2] | AL[2] | AL[2] |
| Average grain size [$\mu$m] | 22 | 63 | 63 | 63 | 28 | 28 |
| Dosing quantity [mg/m³] | 50 | 50 | 150 | 300 | 150 | 300 |
| Bypass Last opening before measurement | 1 day | 3 days | 4 days | 2 days | - | 1 day |
| Opening time | 86 min | 1 min | 5 min | 2 min | - | 5 min |
| Dust emission [mg/m³] | - | 87 | < detection limit | 4 | < detection limit | 4 |
| Dust type | no measurement | coarse | - | extra-fine dust | | extra-fine dust |

Especially at the start of the tests, difficulties with the process control system of the incineration plant were frequently encountered which resulted in the filtering system having temporarily to work in a bypass. In this case, the dust-laden waste gas is directly conducted to the clean gas pipe where part of the dust particles settle. In the case of a closed bypass, these dust accumulations automatically give rise to increased dust emissions which--depending upon the duration of the preceding bypass operation--will only slowly return to the zero emission value of the filtering system. Due to the frequent and long-lasting bypass operation at the test start, the

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[1] AC stands for activated carbon. [2] AL stands for activated lignite

waste gas pipe downstream of the fabric filter was covered with heavy dust deposits which were only removed in the course of a longer period. Thus, the dust sample of the second test series with a dust concentration of 87 mg/m³ showed easily detectable dust particles with a diameter of up to 1 mm. An adjusted operation permitted a considerable reduction in the bypass frequency during the further investigation. Hence, the dust contents in the clean gas reveal marked decreases with values of far below 5 mg/m³. As an example of dust released in the clean gas after bypass operation, Fig. 4 shows the course of dust emission directly after the opened bypass.

Fig. 4:    Course of dust emission after bypass operation
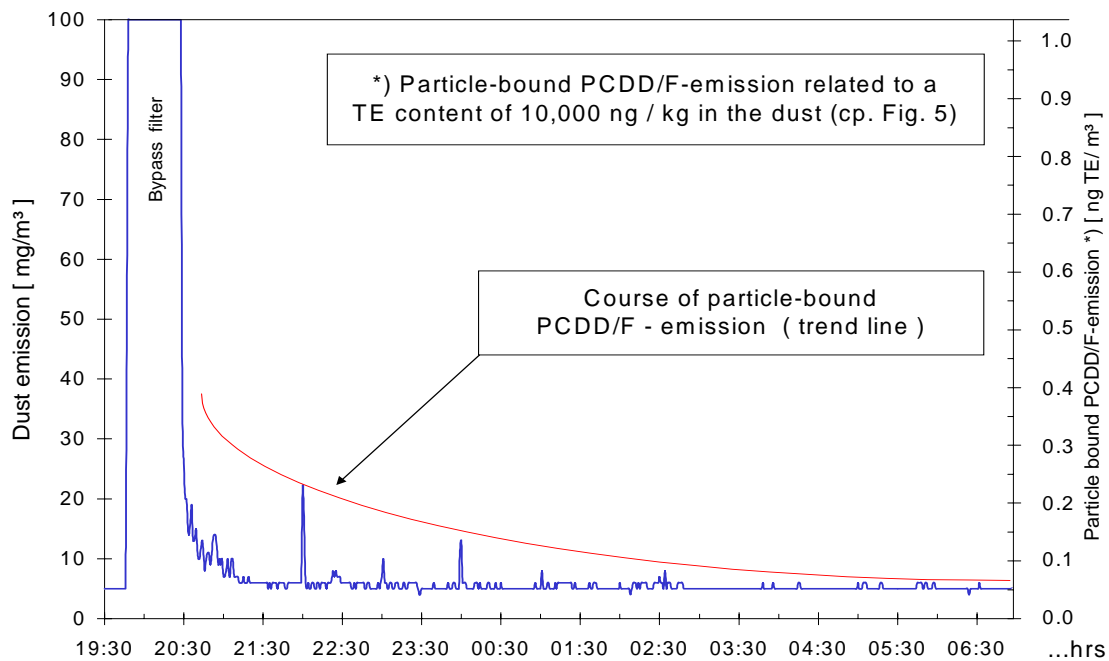


To obtain statements about the quantitative influence exerted by the dust emission on the PCDD/F content, Fig. 5 shows the PCDD/F contents measured in the clean gas during the activated lignite-based tests that are graphically represented with the corresponding carbon concentrations reached in the filter dust (dosing adjustment) and assessed related to the dust contents determined in the respective clean gas.

During the tests with pulverized activated lignite additions (median: 63 $\mu$m) of 150 mg/m³ and 300 mg/m³, any considerable dust emissions (< 4 mg/m³) were not detected in the clean gas so that the obtained PCDD/F clean gas values of 0.19 ng TE/m³ with 150 mg of pulverized activated lignite (63 $\mu$m) per m³ and of 0.12 ng TE/m³ with 300 mg of pulverized activated lignite (63 $\mu$m) per m³ describe more or less the "dust-free" condition. If we assume a linear dependence of the feasible PCDD/F values in the clean gas on the carbon concentration in the filter dust (dosing rate), we obtain a theoretically feasible dust-free PCDD/F value of 0.23 ng TE/m³ in the case of a pulverized activated lignite addition of 50 mg/m³. The difference from the actually measured cumulative value of 1.13 ng TE/m³ (dusty and gaseous dioxins and furans) yields a particle-bound PCDD/F share of 0.90 ng TE/m³. If we

relate this TE value to the average dust content of 87 mg/m³ recorded in the clean gas at the time of measurement, we arrive at a dioxin load of approx. 10,000 ng TE/kg in the dust. The loading is in the range of the TE contents in the filter dust that is usual for household refuse incineration plants. From the level of the dioxin load in the filter dust it can be derived that especially in household refuse incineration plants the proper functioning of dust separation is--in addition to adsorption--a major element of PCDD/F reduction and can exert a decisive influence on the success of process-integrated adsorption.

Fig. 5:     Assessment of the PCDD/F load in the filter dust
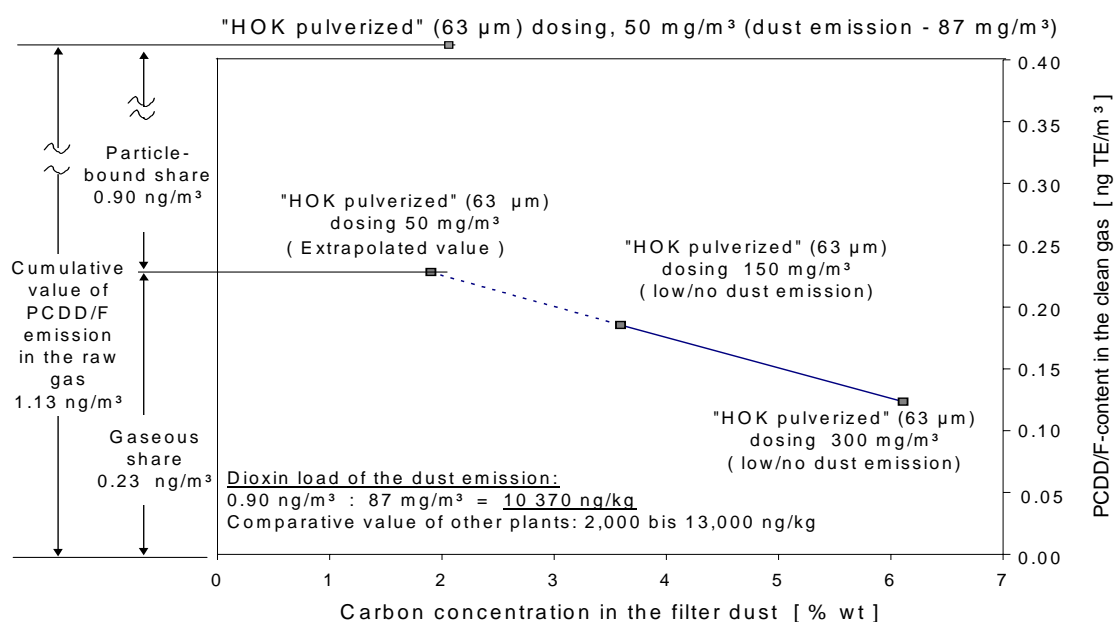


## Influence of Grain-Size Quality and Sorbent Dosing on PCDD/F Reduction

Fig. 6 shows the dependence of the PCDD/F reduction on the carbon concentration in the filter dust (dosing rate) and the grain size of the activated lignite used.

As shown in Fig. 6, the PCDD/F reduction in the case of activated lignite (63 $\mu$m) and reactivity-enhanced pulverized activated lignite (28 $\mu$m) is considerably stepped up with increasing carbon content in the filter dust. From the above influence of the carbon concentration in the filter dust on the obtainable clean gas value it can be derived that the dust occurring in the raw gas at a constant dosing rate exerts--due to the dilution effect--an appreciable influence on the obtainable clean gas value. This in turn demonstrates that the dosing rate required for efficient pollutant separation is dependent on the process dust and thus specific to the plant, and not--as unfortunately done in too many cases--transferrable to different plants as design value. In the present case, and in order to obtain optimum conditions for pollutant reduction, the values determined should not fall below a carbon concentration of 6 % wt in the filter dust in the case of pulverized activated lignite

use (63 μm) and of 3 % wt in the case of reactivity-enhanced pulverized activated lignite use (28 μm). The differences in the carbon contents detected in the filter dust in the case of pulverized activated lignite and reactivity-enhanced pulverized activated lignite additions at the same dosing rate are due to the process and result from fluctuations of the dust concentration in the raw gas.

Fig. 6: Dependence of PCDD/F reduction on the sorbent amount and the grain size
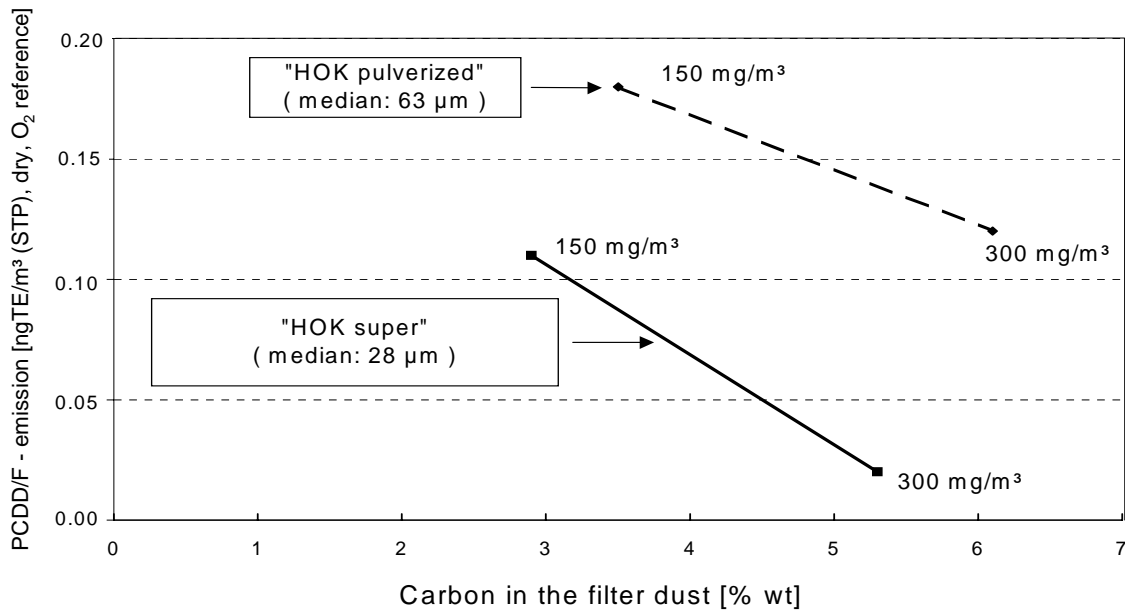


The comparison of the results obtained with different grain-size qualities clearly shows that with a stepped-up milling rate the adsorptive capacity can be increased considerably. So, compared to pulverized activated lignite ("HOK pulverized") the use of reactivity-enhanced pulverized activated lignite ("HOK super") at the same dosing rate allowed the separation efficiency to be more than doubled. Thanks to the use of "HOK super", the PCDD reduction was increased to such a degree that a clean gas value of far below 0.1 ng TE/m³ was reached.

In order to examine the continuous sorbent addition, the outlet of the dosing equipment was provided with a radar motion detector for the tests to be carried out which works according to the Doppler's principle and continuously records the measuring signal. As an example of the logs recorded, Fig. 7 shows a typical curve.

The section "dosing failure" describes the condition of interrupted sorbent dosing. Planned dosing interruptions occurred in the case of turned-off lime milk dosing and opened bypass. This showed that the controlling chain was not equipped with an automatic restarting mechanism. This in turn resulted in the restart of the dosing mechanism--which was to be manually performed by the operating personnel--being temporarily omitted. In addition, some rather long dosing failures were detected at irregular intervals which were caused by an unsuitably positioned min. probe (see Fig. 8). "Insufficient dosing" was observed at irregular intervals in all test configurations. This was due to discharging problems in the receiving bin which

were largely remedied by the cyclic moving of the receiving bin in the measuring period of PCDD/F sampling. The technical measure envisaged is the installation of a rapper.

Fig. 7.     Course of dosing control



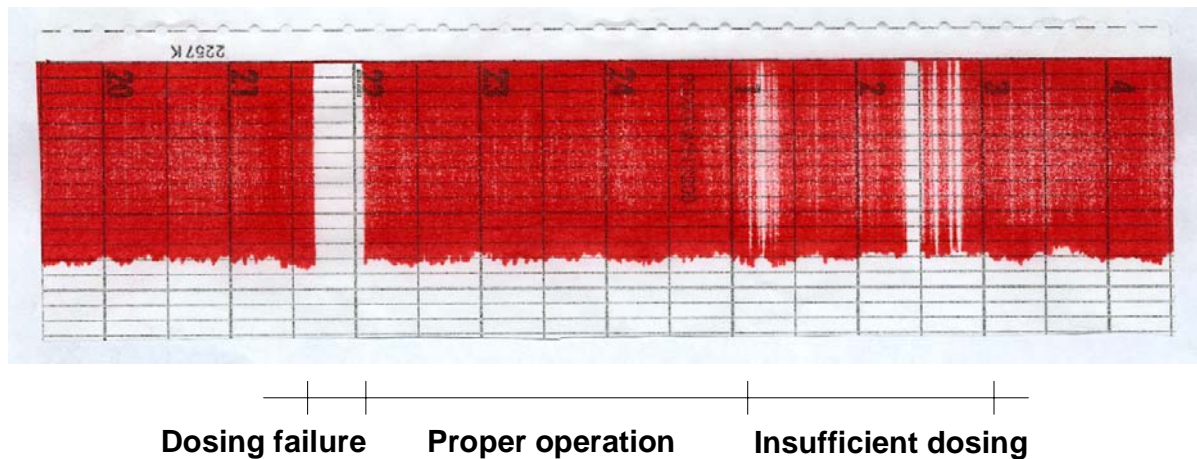**Dosing failure     Proper operation     Insufficient dosing**
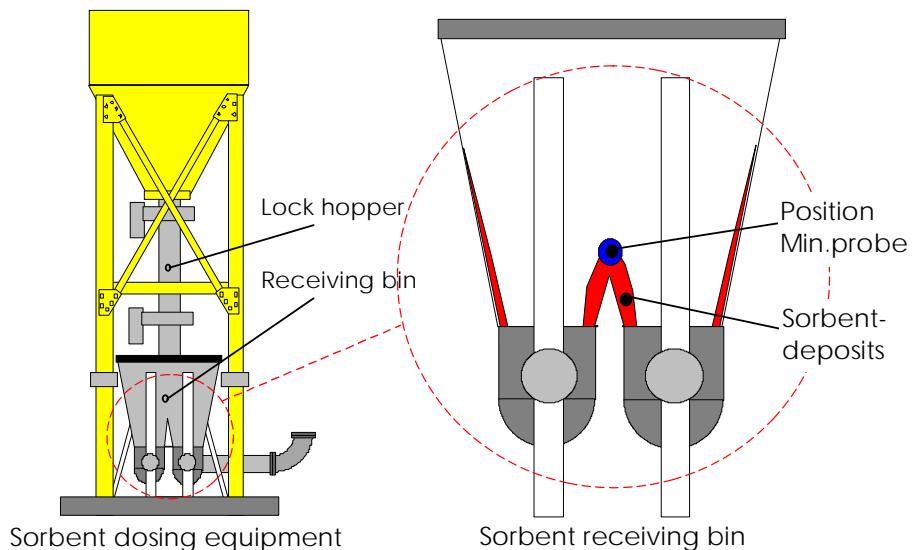
Fig. 8:     Sorbent receiving bin



## Influence of Carbon Distribution in the Filter on PCDD/F Reduction

An important prerequisite for efficient pollutant reduction in the process-integrated adsorption according to the filter-bed process is the uniform distribution of the

sorbent in the fabric filter and the prevention of major non-uniform concentration distribution.

For this reason, dust samples were taken from the separated filter dust at the existing discharging cones of the filtering system during each measuring series and then analysed for their carbon contents. The schematic filter set-up is shown in Fig. 9. Sampling took place at the existing eight discharging cones of the filtering system.

Fig. 9:     Filter set-up



Lateral view:

Outlet pipe

Filter 5    Filter 6    Filter 7    Filter 8

Inlet pipe

Discharge through filter cones

Topview:

Filter-section 1    Filter-section 2    Filter-section 3    Filter-section 4

Inlet pipe lying underneath
Outlet pipe lying above

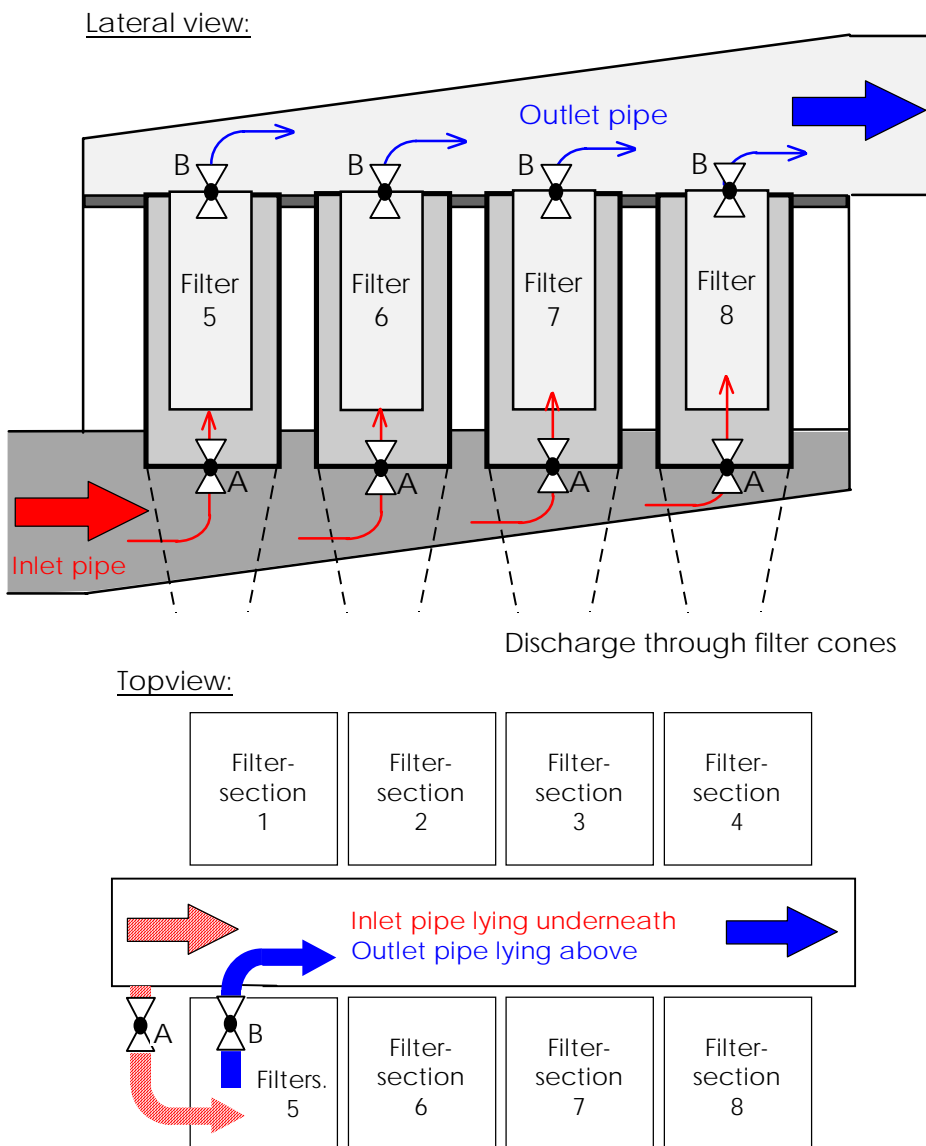Filters. 5    Filter-section 6    Filter-section 7    Filter-section 8

Table 3 lists the carbon contents analysed from the filter dust during the different test- series.

For all test configurations, the carbon concentrations detected in the filter dust samples show a uniform distribution over the filter. No unacceptable non-uniform concentration distributions, which partly limit the adsorptive effect, were found. As expected, the carbon content rises with the dosing rate.

Table 3:    Carbon concentration in the filter dust

| Test series | | 1 | 2 | 3 | 4 | 5 | 6 |
|---|---|---|---|---|---|---|---|
| Adsorbent | | AC[3] | AL[4] | AL[4] | AL[4] | AL[4] | AL[4] |
| Average grain size | [μm] | 22 | 63 | 63 | 63 | 28 | 28 |
| Dosing quantity | [mg/m³] | 50 | 50 | 150 | 300 | 150 | 300 |
| Carbon content in the filter dust | | | | | | | |
| Discharging cone 1 | [% wt] | - | 1.9 | 3.1 | 6.1 | 2.8 | 5.4 |
| Discharging cone 2 | [% wt] | - | 1.8 | 3.8 | 6.6 | 3.1 | 5.1 |
| Discharging cone 3 | [% wt] | 1.5 | 2.0 | 3.5 | 5.8 | 3.0 | 5.5 |
| Discharging cone 4 | [% wt] | 2.0 | 1.9 | 3.3 | 5.7 | 3.3 | 5.5 |
| Discharging cone 5 | [% wt] | 1.9 | 2.3 | 3.8 | 6.0 | 2.7 | 5.2 |
| Discharging cone 6 | [% wt] | 1.9 | 2.0 | 4.1 | 7.1 | 2.7 | 5.1 |
| Discharging cone 7 | [% wt] | 1.8 | 2.1 | 3.7 | 5.8 | 2.5 | 5.3 |
| Discharging cone 8 | [% wt] | 1.8 | 1.8 | 3.5 | 5.8 | 2.8 | 5.2 |
| Mean value Cones 1-8 | [% wt] | 1.8 | 2.0 | 3.6 | 6.1 | 2.9 | 5.3 |

## CONCLUSION

As shown by the results obtained from the operational tests in a refuse incineration plant for municipal and industrial wastes, it is feasible to use activated lignite for pollutant separation with high separating efficiencies in a plant designed for activated carbon use, without any modifications to the existing infrastructure being required. The results attained at different dosing rates and with various grain-size qualities reflect the influence on the obtainable PCDD/F reduction. The high cleaning efficiency of activated lignite shown by the results from dioxin reduction becomes all the more obvious if we consider that due to the existing equipment the tests were run under unfavourable conditions. Despite a non-optimum plant situation, a clean gas value of 0.02 ng TE/m³ was obtained in the case of reactivity-enhanced pulverized activated lignite ("HOK super") addition, which thus remained far below the limit value of 0.1 ng TE/m³ applicable to refuse incineration plants. The assessment of the PCDD/F emission in connection with the dust emission shows that the emitted particle-bound dioxins and furans exert an appreciable influence on the PCDD/F emission level.

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[3] 'AC' stands for activated carbon.
[4] 'AL' stands for activated lignite.

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