

Significance of and Challenges for Flue Gas Treatment Systems in Waste Incineration

Rudi Karpf and Andreas Wiedl

1.	Significance and development of flue gas treatment in Germany and in Europe since 1970
1.1.	History
1.2.	Development of legal emission regulations on the example of German law.....
2.	Challenges for flue gas treatment systems in the future.....
3.	References

Flue gas cleaning downstream of waste incineration plants had its origins in the increased construction and deployment of such plants to counter rising air pollution in the nineteen-sixties. Back then, the ever-growing burden on the environment caused lawmakers to start enacting emission limits for air pollution control. An unceasing series of environmental scandals and increasingly better analytical methods and measuring instrumentation led to a constant reduction of the emission limits and, consequently, to ongoing adjustment and further development of the necessary process stages in flue gas cleaning. As a result, today minimum emissions can be reached even under the challenging condition of deployment of a very inhomogeneous fuel (waste) and, hence, waste incineration today is no longer a key contributor to air pollution. Today, the need for flue gas cleaning is not called into doubt anymore and has long become a matter of course in the industry and in society at large. Apart from ensuring efficient elimination of noxious gases, the focus of today's further developments is on issues such as energy efficiency, minimization of input materials and recovery and recycling of by-products from flue gas cleaning as valuable raw materials. These issues are also deemed to be key challenges, especially when it comes to selecting sites for new plants in such a manner that potential synergies can be exploited. Such aspects will also have to be considered in the plans for the predicted *mega-cities* of the future.

1. Significance and development of flue gas treatment in Germany and in Europe since 1970

1.1. History

The necessity and relevance of cleaning pollutant-laden exhaust gas flows can be traced back to the 16th century. The 19th century, then, saw the emergence of more comprehensive and sophisticated cleaning measures as industrialization took its course.

The first commercial electrostatic precipitator mentioned in literature, for instance, was commissioned in 1885 by Walker and Hutchings at the tail end of a lead melting plant in Northern Wales [10]. In the following years, F.G. Cottrell in the United States made significant contributions to the development of filter technologies, especially for filter technology deployed in the metallurgical industry. In Europe, too, development of flue gas cleaning technologies continued, as air pollution and its impact on human health, caused by the ever increasing number of factories, thermal production processes and power plants became more and more noticeable and notorious.

The first waste incineration plant on the European mainland was built and commissioned in 1896 in Hamburg-Bullerdeich for hygienic reasons (Figure 1), while in London, the first waste incineration had been commissioned as early as in 1870. However, apart from fly ash removal making use of the mass inertia effect by suitably arranging exhaust gas ducts and baffles, no other flue gas cleaning efforts were made in such plants.

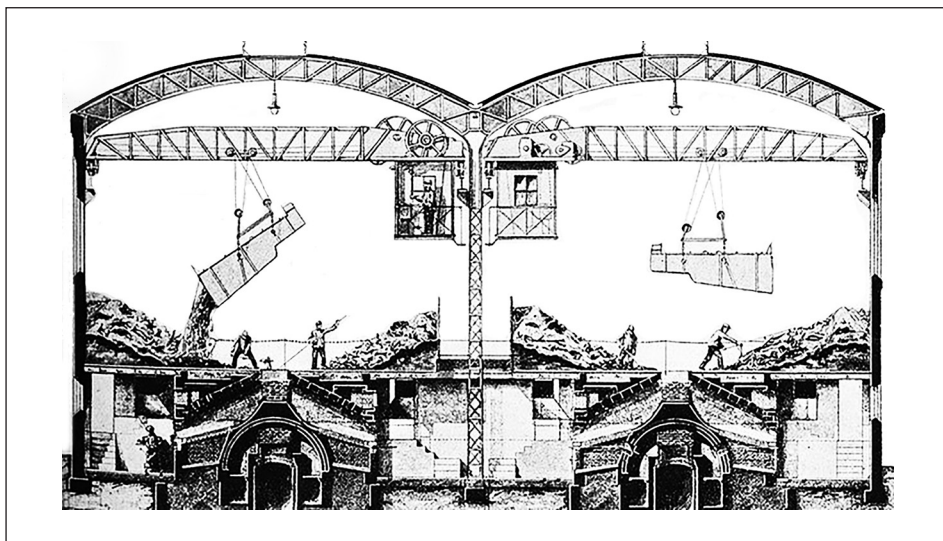


Figure 1: First waste to energy facility in Germany, Hamburg-Bullerdeich

Source: Dietrich, O.; Die Stadt, der Müll und der Schiet; www.NDR.de/Kultur/Geschichte, 10.09.2012

In Germany, waste incineration acquired greater significance not before the second half of the 20th century. Due to the rapid economic growth after World War II, the waste volumes to be managed and disposed of grew in an unparalleled way.

1.2. Development of legal emission regulations on the example of German law

Against the background of increasing air pollution, lawmakers made repeated but still occasional efforts to reduce air pollution by enacting legal regulations. The first systematic corpus of air pollution control rules and regulations in Germany finally

Table 1: Development of the legal emission regulation from 1974 up to now

Pollutant	Unit	TA Luft 1974 version	TA Luft 1986 version	17. BImSchV 2003 version	EU Guideline 2000/76		13. BImSchV 2004 version 50 MW < RTI < 100 MW	TA Luft 2002 version RTI < 50 MW	17. BImSchV 2013 version		
		General requirements	General requirements	DAV	DAV	HHAV	DAV	General requirements	HHAV	DAV	YAV
O ₂ -reference percentage	Vol.-% dry	11	11	11	11	11	6	7 – 11		11	
Dust	mg/m ³	100	30	10	10	30	20	20	20	5 (10 for RTI < 50 MW)	
Total Organic Carbon (TOC)	mg/m ³	-	20	10	10	20	-	50			
Hydrogen chloride (HCl)	mg/m ³	100	50	10	10	60	-	30	60	10	
Hydrogen fluoride (HF)	mg/m ³	5	2	1	1	4	-	3			
Carbon monoxide (CO)	mg/m ³	1,000	100	50	50	100	150	150			
Sulphur dioxide (SO ₂)	mg/m ³	-	100	50	50	200	850	350 – 1,300			
Nitrogen oxide (NO ₂)	mg/m ³	-	500	200	200**	400	400	150 – 500	400	150 (200 for RTI < 50 MW) ⁽⁵⁾	100 ⁽⁵⁾ (7)
Ammonia (NH ₃)	mg/m ³	-	-	-	-	-	-	-	15	10	-
Heavy metals	mg/m ³	-	-	0.03	0.05	0.05	0.03	0.05	0.05	0.03	0.01 ⁽⁵⁾ (6)
Mercury (Hg)	mg/m ³	-	-	0.03	0.05	0.05	0.03	0.05	0.05	0.03	0.01 ⁽⁵⁾ (6)
Dioxins and furans	ng/m ³	-	-	0.1	0.1	0.1	0.1	-	-	-	-
Class I	mg/m ³	20***	0.2	0.05	0.05	0.05	0.05*	0.05 ⁽²⁾	-	0.1	-
Class II	mg/m ³	50***	1	0.5	0.5 ⁽¹⁾	0.5 ⁽¹⁾	0.5*	0.5 ⁽³⁾	-	0.5	-
Class III	mg/m ³	75***	5	0.05	-	-	0.05*	1 ⁽⁴⁾	-	0.05	-

The concentration data is based on standard temperature and pressure, dry state, for each oxygen reference value; DAV indicates daily average value; HHAV indicates half hourly average value; YAV indicates yearly average value; Heavy metals class I: Σ Sb, As, Pb, Cr, Co, Ni, Cu, Mn, V, Sn; Heavy metals class II: Σ Sb, As, Pb, Cr, Co, Ni, Cu, Mn, V, Sn; Heavy metals class III: Σ As, benzopyrene, Cd, Co(aq), Cr(IV)

* not applicable to use of coal, untreated wood only; ** combustion capacity > 6t/h or new facilities; *** related to the former classification

⁽¹⁾ excluding Sn; ⁽²⁾ applicable to TI (single substance); ⁽³⁾ applicable to Pb, Co, Ni, Se, Te; ⁽⁴⁾ applicable to Sb, Cr, CN, F, Cu, Mn, V, Sn; ⁽⁵⁾ not applicable to use of existing plants with RTI < 50 MW; ⁽⁶⁾ to be valid as of 2019; ⁽⁷⁾ not applicable for existing plants; ⁽⁸⁾ applicable to Mercury if the emission value is always < 20 % of the requested emission value

RTI: Rated Thermal Input

came into effect in 1964, in the form of the German clean air directive *Technische Anleitung zur Reinhaltung der Luft*, referred to as *TA Luft* in short. In the years that followed, the *TA Luft* directive was revised several times and in 1974 it was re-enacted as the First General Administrative Regulation to the Federal Immission Control Act (*Erste Allgemeine Verwaltungsvorschrift zum Bundesimmissionsschutzgesetz*) [6].

Table 1 shows the development of the statutory emission limits until today. The emission limits stipulated in the 1974 version of *TA Luft* could be met using a dust removal stage, usually in the form of an electrostatic precipitator, and by adding dry hydrated lime for reduction of hydrochloric acid (HCl) if necessary. No limit was stipulated for sulphur dioxide (SO_2) at that time.

The *Ordinance on large combustion plants* (*Verordnung für Großfeuerungsanlagen*, 13th BImSchV) adopted in 1983 reflected the political response to increased occurrence to winter smog in major German cities and the widespread phenomenon of dying forests caused by so-called acid rain. The 13th BImSchV left its mark on the 1986 amendment of *TA Luft*, which back then still served as the basis for the definition of emission limits for waste incineration plants. As a result of the 13th BImSchV, the emission limits for particulate matter, carbon monoxide (CO), hydrochloric acid (HCl) and hydrofluoric acid (HF) were drastically reduced. For the first time, emission limits were set for sulphur dioxide (SO_2) and nitrogen oxides (NO_x), and the limits for heavy metals of the different classes were lowered by up to three powers of ten. Furthermore, the general grandfather clause for existing plants was abolished with the amendment. This meant that existing flue gas cleaning stages had to be expanded or modified. As shown in Figure 2 to 4, frequently a wet scrubber was added downstream of the dust removal stage (electrostatic precipitator), or the electrostatic precipitator was replaced by a conditioned dry sorption stage with a fabric filter. The conditioned dry sorption system was either designed as a spray adsorption system (spray injection of lime slurry) or it included an evaporation cooler and addition of dry hydrated lime.

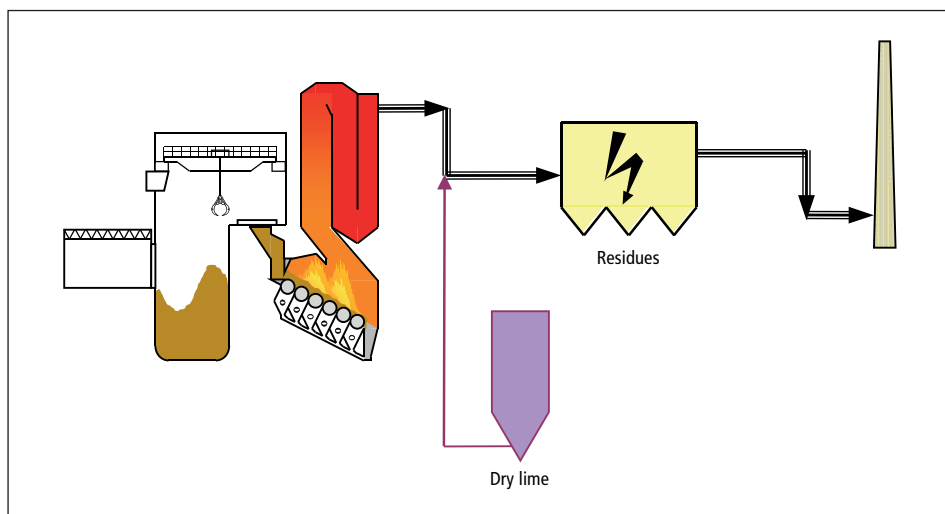


Figure 2: Flue gas treatment according *TA Luft* 1974 version

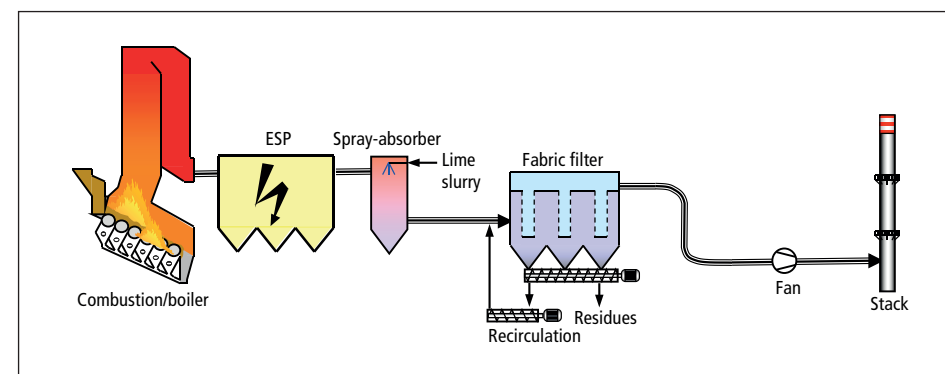


Figure 3: Flue gas treatment according *TA Luft* 1986 version, spray absorption system

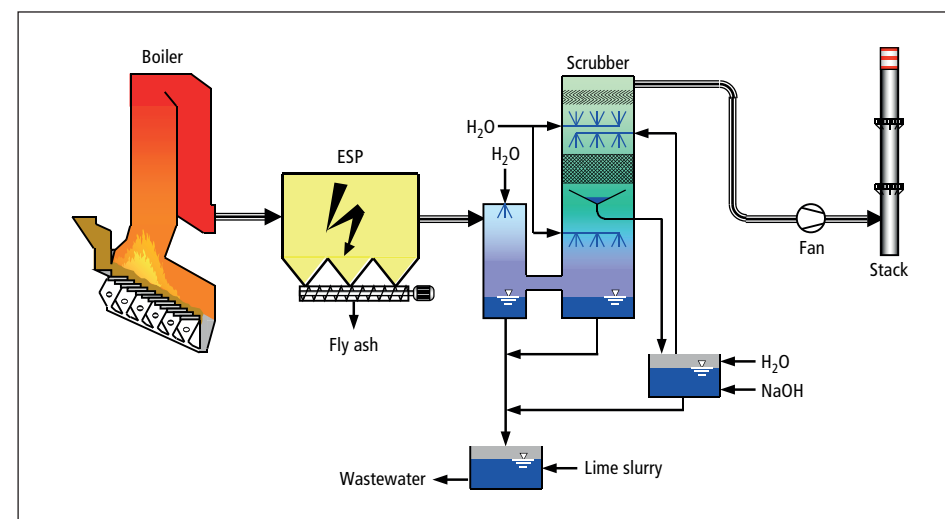


Figure 4: Flue gas treatment according *TA Luft* 1986 version, wet scrubber system

In the aftermath of the Seveso dioxin disaster and the constantly improving analytical and measurement methods, also the waste incineration plants moved into the focus of public attention as sources of dioxin emissions. This caused lawmakers to further tighten the emission limits for waste incineration plants. The new limits were no longer governed by the *TA Luft* directive, which until then had also been applicable to waste incineration plants, but instead in a separate ordinance on the implementation of the Federal Immission Control Act dedicated especially to waste incineration and co-incineration (17th BImSchV). This new directive governing waste incineration stipulated another significant reduction of the limit values applicable until then. Furthermore, due to its high toxicity, special requirements were stipulated for mercury (Hg) as a chemical element from the group of heavy metals, and an emission limit of 0.1 ng/m^3 TE for dioxins/furans (PCDD/PCDF) of 0.1 ng/m^3 TE was stipulated for the first time. As a result, all existing plants and new plants to be built were to be provided with a

tail-end nitrogen oxide reduction system and an advanced Hg and PCDD/PCDF reduction stage. For nitrogen oxide removal (deNO_x), two processes have found general acceptance: the selective non-catalytic reduction (SNCR) process and the selective catalytic reduction (SCR) process. Removal of Hg and dioxins was mostly achieved by means of an adsorptive process using carbon particles, in the form of moving bed or entrained flow adsorbers (fabric filters). In some cases, Hg removal was achieved by application of a chemisorptive process in a wet scrubber or in an adsorptive process using zeolites. Catalytic oxidation of dioxin was also an option for dioxin removal in plants where a catalytic NO_x reduction system was deployed. Within the scope of such retrofitting efforts, some flue gas treatment systems were installed which were designed to minimize the amount of non-recyclable residues produced in the flue gas treatment process. As a result, hydrochloric acid rectification and gypsum producing systems were added in the nineteen-nineties to selective treatment stages such as wet scrubbers. Also refer to Figure 5.

As it turned out quickly that there was no market in Germany for the by-products from flue gas treatment systems of waste incineration plants, and the energy expenditure and plant construction effort needed for production of the by-products was very high, such add-on process stages did not have much of a future. In the same period, while these insights were gained, waste incineration underwent a metamorphosis from mere waste disposal to energy recovery from waste. From then on, energy efficiency was in the focus and became an important design criterion both for new and modified plants. Until today, the energy efficiency of waste incineration plants is assessed using the *political* energy efficiency factor $R1$, and plants are accordingly classified as either *Disposal operations* or *Recovery operations*. Incineration facilities for solid municipal waste are classified as *Recovery operations* if their $R1$ factor is at least 0.60 or 0.65 (depending on whether the permit was issued before 01 January 2009 / after 31 December 2008 or not).

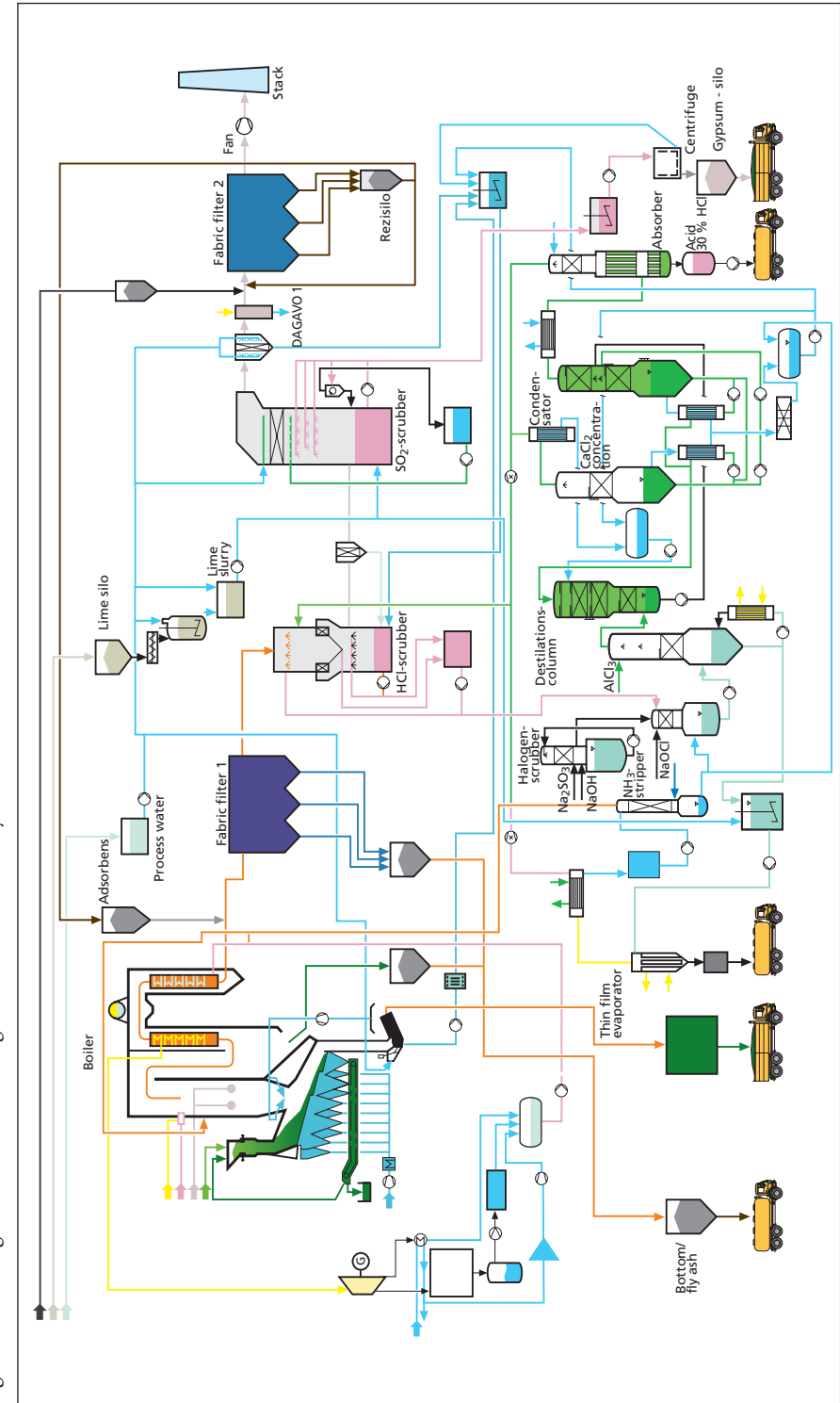
The emission limits were last tightened in the amendment of the 17th *BImSchV* enacted in May 2013: Under the pressure of rising respiratory dust levels and nitrogen oxide emissions, the limits for these pollutants have been reduced, while a new limit has been stipulated for ammonia emissions for the first time. Furthermore, the emission limit for mercury was tightened within the scope of international mercury reduction policies based on the Minamata follow-up conference.

This means that future challenges for flue gas cleaning will continue to be high pollutant removal efficiency and very high efficiency in the use of energy and consumables (minimum consumption of resources).

2. Challenges for flue gas treatment systems in the future

As described above, present-day flue gas cleaning systems are unique in design and their specific configuration often reflects the development of the emission limits over time. Emission monitoring ensures that emissions from all plants in service today stay safely below the statutory limits. However, as a tightened NO_x emission limit must be complied with from 2019 onwards while compliance with a defined NH_3 emission

Figure 5: Flue gas treatment according 17. BImSchV with recyclable fraction extraction



Source: Thomé-Kozmiensky, Elisabeth (Ed.), Abfallverbrennungsanlagen – Deutschland – 2014 | 2015; Neuruppin: TK Verlag, Karl Thomé-Kozmiensky, 2016, S. 282

limit must also be ensured, most of the plants equipped with a selective non-catalytic (SNCR) deNO_x stage will need optimization or adjustment. Depending on the design and scope of plant and equipment of the SNCR it will in many cases be impossible to achieve NO_x emission levels of less than 150 mg/m³ while at the same time complying with an NH₃ emission limit of < 10 mg/m³.

This may be due to the following root causes, identification of which can also point the way to possible optimization measures:

- insufficient spray injection points for ammonia water (NH₄OH) or urea (CH₄N₂O) as reducing agents;
- possibilities to switch between different injection lances and control the lances individually;
- quality and scope of temperature measurement;
- knowledge of temperature distribution in the furnace and combustion control;
- insufficient or no feedback on NO_x/NH₃ concentrations downstream of the boiler.

In consideration of the root causes above describe, the following approaches to finding a solution are conceivable and should be mentioned here together with experience already gained. Optimized operation of an SNCR system fundamentally requires a dynamic detection of the furnace temperature, ideally with several measurement sections across the furnace cross section, such that temperature imbalances can also be detected. Acoustic measurement techniques and IR¹ pyrometers have proven to be particularly well suited for this purpose. On the basis of the temperature distribution so ascertained and using suitable spray injection equipment, the reducing agent can then be variably injected into the temperature zone that is most conducive to NO_x reduction. Such selective spray injection can be accomplished by either providing several levels of injection nozzles and changing over between these levels as appropriate or by using swivelling injection lances. For the SNCR system to be controlled in the best possible manner, it is also an advantage if the parameters NO_x and NH₃ can be measured by means of dedicated process instrumentation directly after the furnace instead of having to do with the signals from the emissions measurement system arriving with some delay.

Another option for optimization addressing in particular the issue of ammonia slip is the deployment of an additional catalytic treatment stage which permits a subsequent reaction of the slipped NH₃ with the NO_x still present in the flue gas. Such a slip catalyst can be arranged in the boiler (temperature range of approx. 280 to 300 °C), although exposure to the untreated flue gas makes special demands on the design of the catalyst; especially catalyst fouling and clogging due to the high fly ash concentration should be mentioned in this regard.

If the flue gas cleaning process includes a fabric filter which is operated at a temperature of not less than 180 °C, this filter can be equipped with catalytic filter bags, in which the catalytic medium is protected against fouling by an upstream diaphragm. The actual function of the fabric filter as a dust removal and sorption stage can thereby expanded to also perform the function of NH₃ slip reduction or even serve as a deNO_x stage.

¹ IR = infrared

Increasing energy efficiency will continue to be another challenge for flue gas treatment processes. A very comprehensive and detailed study [4] on the net emissions balance as a function of energy expenditure shows that the energy expenditure for a multi-stage flue gas treatment system with minimum emissions is not necessarily higher than that of single-stage systems. Modern-day and future know-how regarding the design of efficient plants will not necessarily imply the development of new processes, but rather place a focus on the intelligent combination and configuration of proven process stages. One example of this is the Delfzijl waste incineration facility in the Netherlands.

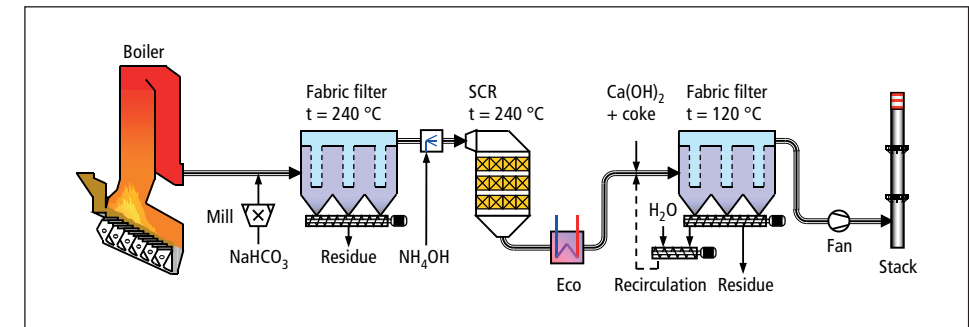


Figure 6: Flue gas treatment of the waste to energy plant Delfzijl (NL)

Source: Karpf, R.; Emissions-related energy indicators for flue gas treatment systems in waste incineration; ISBN 978-3-944310-14-5, TK Verlag Karl Thomé-Kozmiensky, Neuruppin 2014

As regards the future selection of sites for new plants, this implies that plants should be built at sites where a suitable infrastructure including energy sinks exist. Owing to very effective multi-stage flue gas treatment, the plants do not place a burden on the environment, even if they are built within cities or metropolises. Another option is a siting approach similar to that currently practiced in Copenhagen, the capital of Denmark. There, the entire design of a waste-to-energy plant, including use of the energy produced, is matched to the purpose of a leisure facility project developed in parallel. There, too, minimum emission levels are ensured by deployment of an effective multi-stage flue gas treatment system in order to avoid that any burden is placed on the environment.

In most cases, the intelligent combination and configuration of process stages for energy optimization also affords the benefit of achieving the best possible process conditions and thus maximum pollutant removal efficiency and minimum consumables consumption.

The most recent example is the publication of newly developed deNO_x catalysts which, designed as low-temperature catalysts for the first time achieve serviceability in temperature ranges < 150 °C, thereby enabling application of an upstream, lime-based dry sorption process without additional heating, which was up to now the sole domain of processes using sodium bicarbonate.

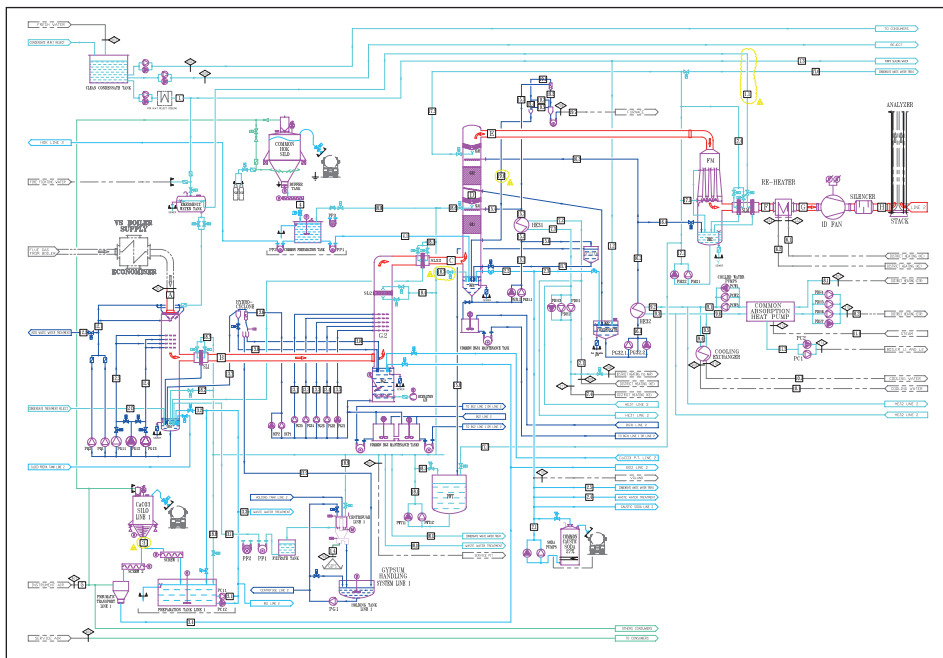


Figure 7: Waste to energy facility Copenhagen, Denmark

Source: Fuchs, Ch.; Gestern Müllverbrennungsanlage! Heute Freizeitpark? Das Konzept der Amagerforbrænding in Kopenhagen; 12. Potsdamer Fachtagung - Optimierungen in der thermischen Abfall- und Reststoffbehandlung Perspektiven und Möglichkeiten; Potsdam 26. - 27. Februar 2015

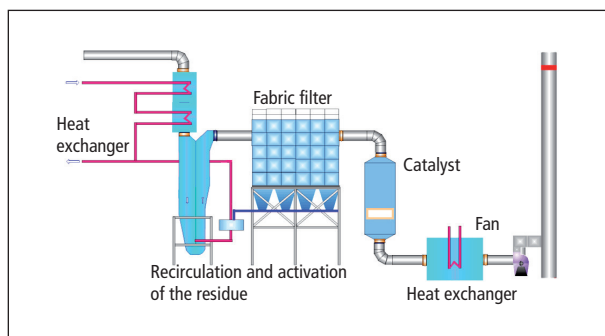


Figure 8:

VapoLAB and CataLABVLT

Sources:

Paulet, Ch.; Tabaries, F., et. al.; Eine neue Generation der Very Low Temperatur SCR für Abfallverbrennungsanlagen – Erste Erfahrungen bei Betriebstemperaturen von 140 °C – Thomé-Kozmiensky, K. J.; Beckmann, M. (Eds.): Energie aus Abfall, Band 13. Neuruppin: TK Verlag Karl Thomé-Kozmiensky, 2016

Apart from energy efficiency, recent discussions indicate the imminent renaissance of recovery of valuable by-products from the flue gas – this time, however, not in the form of producing materials such as gypsum or hydrochloric acid, but in the form of feeding the combustion product CO_2 back into the carbon cycle, for instance in the form of methanol. Such considerations have been triggered by political targets as regards climate protection and decarbonization of future energy supply at large.

Synthesis of methanol from CO_2 may be a sensible option for some waste incineration plant sites lacking other options for utilization of the energy due to the nature of the nearby infrastructure. A part of the energy required for the methanol synthesis and

for electrolytic production of hydrogen could then be provided by excess thermal and electrical energy from the waste incineration process. However, due to the relatively low electrical net output of such plants, complete coverage of the electrical energy demand for methanol synthesis by the output from the plant itself will only be possible if just a small part of the total CO_2 output is captured and processed.

In reverse, this means that it will be necessary to import electrical energy from the public grid if almost complete CO_2 capture (ninety percent capturing efficiency) is to be achieved. In this context, the question arises, whether and to what extent this can be used for storage of electricity from fluctuating renewable energy sources. However, in order to be able to perform the function of a storage system for excess electrical energy, additional components for compression and storage of electrolytically produced hydrogen and of carbon dioxide captured from the flue gas flow will need to be provided. Further details can be found in [5].

As already mentioned above in the context of energy efficiency, it is not important to develop entirely new systems or processes, but rather to combine available processes in an intelligent manner and exploit synergies. In this spirit, a symbiosis can be established between the capture and provision of CO_2 (valuable resource) from the flue gas flow and the use of excess electrical energy from fluctuating renewable sources of energy for methanol synthesis.

Another approach for using the CO_2 present in the flue gas was pursued in the energy-from-waste plant in Twence, the Netherlands, where sodium bicarbonate is successfully produced from CO_2 and soda, for use as an additive in flue gas treatment processes. Details on this CO_2 Mineralization Process for sodium bicarbonate production can be found in [3].

In the *ReNaBi* process [7], CO_2 from the flue gas is likewise used to produce sodium bicarbonate in a cyclic process, by recycling residues arising from dry flue gas cleaning. The project is already beyond the pilot plant stage and a first commercial-scale facility is now under construction (to be commissioned in 2017/2018).

3. References

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