Thomas Hilber

UPSWING:
An advanced waste treatment concept compared to state-of-the-art

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UPSWING: An advanced waste treatment concept compared to state-of-the-art

(UPSWING: Ein fortschrittliches Abfallbehandlungskonzept
im Vergleich mit dem Stand der Technik)

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Preface

The present thesis originates from my graduate occupation at the Institute of Process Engineering and Power Plant Technology (IVD) at the University Stuttgart.

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Finally I sincerely thank my family for their support during my studies.

Meiner Mutter

Abstract

The present thesis is set out to assess the potential of a novel and innovative approach for the recovery of energy from municipal solid waste, in the following referred to as the UPSWING concept. This (theoretical) concept describes the coupling of a waste incinerator with a conventional power plant, both on the steam- and on the flue gas side. The waste flue gases are pre-cleaned prior to injection into the power plant process in order to keep substances, potentially harmful for the environment as well as for the power plant process, away from the power plants' boiler system.

The concept proposes significant benefits in terms of efficient waste-to-energy conversion, but also towards the required investment costs, which are expected to be considerably lower compared to a same-sized municipal solid waste incinerator. The demand for such advanced concepts is given by European Legislation, setting strict rules on landfill disposal of untreated municipal solid waste, while promoting more environmentally friendly and more efficient waste treatment concepts under the framework of the Community Waste Strategy COM(96) 399.

In this thesis the environmental, operational and economic performance of the UPSWING concept is evaluated by means of a process comparison with other state-of-the-art concepts. In this context, conventional municipal solid waste incineration and direct co-combustion of waste-derived fuels in thermal power plants have been considered. Environmental and operational aspects were investigated by experimental and theoretical means, while the economical aspects were theoretically assessed by a comparison of the anticipated costs for waste treatment. Following this approach, it was possible to identify benefits respectively deficits of the different waste treatment concepts investigated within the scope of this thesis.

In essence, it was found that the UPSWING process is capable to maintain the high environmental standards of municipal solid waste incineration. In terms of process efficiency and necessary investment costs, significant benefits compared to a standalone waste incinerator were found, while restrictions and potential problems could be determined in terms of waste flue gas and steam integration. Nevertheless, the UPSWING process proposes significant advantages and a large-scale demonstration of the process could be the next step to evaluate its overall capabilities.

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Literature

Nomenclature

$\dot{m}_i^{input}$	Mass input of an element i into the process, kg/h
$\dot{m}_{coal}$	Mass flow of coal, kg/h
$\dot{m}_{SRF}$	Mass flow of SRF, kg/h
$\dot{V}_{MSWI}$	Volume flow of waste flue gases, m³/h
c_i^{coal}	Concentration of an element i in the coal, mg/kg
c_i^{SRF}	Concentration of an element i in the SRF, mg/kg
c_i^{MSWI}	Concentration of an element i in the waste flue gases, mg/m³
$\dot{m}_{ash}$	Total ash input, kg/h
$c_{i,\max}^{ash}$	Maximum concentration of an element i in the ash, mg/kg
R_i^{ash}	Recovery rate of an element i in the ash, %
c_i^{ash}	Measured concentration of an element i in the ash, mg/kg
$c_{i,\max}^{FG}$	Maximum concentration of an element i in the flue gas, mg/m³
c_i^{FG}	Measured concentration of an element i in the flue gas, mg/m³
R_i^{FG}	Recovery rate of an element i in the flue gas, %
$Hg^0(g)$	Fraction of gaseous elemental mercury, %
$HgCl_2(g)$	Fraction of gaseous divalent mercury, %
$Hg(p)$	Fraction of particle-bound mercury, %
e_i	Specific process emissions, g/kWh or kg/MWh
ef_i	Specific emissions factors, %

Indices

input	Input concentration
coal	Index referring to the primary fuel
ash	Index referring to the ash
SRF	Index referring to the secondary fuel
MSWI	Index referring to waste flue gases from MSWI
FG	Index referring to generated flue gases

wf/dry	refers to water free or dry state
waf	refers to water and ash free state
ds	refers to dry substance
(s)	refers to solid form of an element
(l)	refers to liquid form of an element
(g)	refers to gaseous of an element

Abbreviations

AAS	Atomic-adsorption-spectroscopy
APC	Air pollution control
APH	Air preheater
BFB	Bubbling fluidised bed
BFS	Bag filter system
BTS	Electrically heated combustion reactor
CFB	Circulating fluidised bed
CFD	Computerised fluid dynamics
CHP	Combined heat and power
CMM	Continuous mercury measurement
DSD	“Duales System Deutschland” (German recycling concept for packaging residues)
ESP	Electrostatic precipitator
FE	Ferrous metals
FF	Fabric filter (bag filter)
FGD	Flue gas desulphurisation system
GCV	Gross calorific value (higher heating value)
HCF	High-calorific fraction (of waste)
IC	Ion-chromatography
I-TEQ	NATO/CCMS international toxic equivalent (PCDD/F)
KSVA	Semi-technical combustion facility
LOI	Loss on ignition
MBT	Mechanical biological treatment (of MSW)
MFC	Mass flow controller
MSW	Municipal solid waste
MSWI	Municipal solid waste incineration (on the grate)
NCV	Net calorific value (lower heating value)
NF	Non-ferrous metals
NIR	Near infrared detection and separation technology
OFA	Over fire air
PAH	Polyaromatic hydrocarbons
PIC	Products of incomplete combustion
PRR	Paper recycling residues
RDF	Refused derived fuel (former synonym for SRF)
RES	Renewable energy sources
SCNR	Selective non-catalytic reduction
SCR	Selective catalytic reduction
SRF	Solid recovered fuel (produced from non-hazardous waste)

TC	Total carbon
TOC	Total organic carbon
TPP	Thermal power plant
UBC	Unburnt carbon

Chemical species and elements

Hg	Mercury	Cd	Cadmium	Zn	Zinc
Tl	Thallium	As	Arsenic	Sb	Antimony
Pb	Lead	Cu	Copper	Ni	Nickel
Fe	Iron	Mn	Manganese	Ti	Titanium
Cl	Chlorine	F	Fluorine	Br	Bromine

CO	Carbon monoxide
CO_2	Carbon dioxide
HCl	Hydrogen chloride
HF	Hydrogen fluoride
NO_x	Nitrogen oxides (sum of NO, NO_2, and N_2O)
SO_2	Sulphurdioxide

HCB	Hexachlorobenzene (C_6Cl_6)
PCP	Pentachlorophenol (C_6HCl_5O)
PCB	Polychlorinated biphenyls ($C_{12}H_{10-x}Cl_x$)
PCDD	Polychlorinated dibenzodioxins
PCDF	Polychlorinated dibenzofurans

PE	Polyethylene
PP	Polypropylene
PS	Polystyrol

Deutsche Zusammenfassung

Das Ziel der vorliegenden Arbeit ist es, das technische Potential eines neuen und innovativen Verfahrens zur energetischen Verwertung von Abfällen zu untersuchen. Das Verfahren selber, im Folgenden als UPSWING Prozess bezeichnet, wurde vom Forschungszentrum Karlsruhe entwickelt und patentiert. Die Ergebnisse dieser Arbeit basieren auf den Untersuchungen, die von mir während meiner wissenschaftlichen Tätigkeit am Institut für Verfahrentechnik und Dampfkesselwesen im Rahmen des von der Europäischen Gemeinschaft geförderten Forschungsvorhabens UPSWING (ENK5-CT-2002-00697) durchgeführt wurden.

Der UPSWING Prozess thematisiert die Kopplung einer Rostfeuerung mit einem konventionellen thermischen Kraftwerk auf Braun- bzw. Steinkohlebasis. Dabei erfolgt die Kopplung beider Einzelprozesse sowohl auf der Rauchgas- als auch auf der Dampfseite. Der im Dampferzeuger der Rostfeuerung erzeugte Prozessdampf wird in den Dampferzeuger des Kraftwerks eingekoppelt und dort, nach entsprechender Überhitzung, im Turbosatz entspannt. Das Kondensat wird entsprechend zurückgeführt. Die Rauchgase der Rostfeuerung werden vor dem Einbringen in den Kraftprozess teilweise gereinigt, um aus umwelt- und verfahrenstechnischer Sicht potentiell kritische Substanzen abzuscheiden. Dabei ist ein wesentliches Ziel des Verfahrens, die hohen ökologischen Standards einer konventionellen Müllverbrennung beizubehalten, gleichzeitig aber die Effizienz der energetischen Verwertung zu optimieren.

Das UPSWING Verfahren verspricht eine signifikante Wirkungsgradsteigerung bei der Konversion der im Abfall gebundenen Restenergie in nutzbare Wärme bzw. Strom. Im Vergleich zu einer konventionellen Müllverbrennungsanlage ergeben sich weiterhin deutlich geringere Investitionskosten. Im Rahmen dieser Arbeit wurde das verfahrenstechnische, ökologische und ökonomische Potential des Prozesses mit Verfahren verglichen, die dem momentanen Stand der Technik entsprechen. Für diesen Vergleich wurden die konventionelle Müllverbrennung sowie die direkte Mitverbrennung von Sekundärbrennstoffen herangezogen. Ökologische und verfahrenstechnische Aspekte wurden, gestützt auf experimentellen und theoretischen Ergebnissen, ausführlich untersucht. Das ökonomische Potential wurde durch einen Vergleich der zu erwartenden Entsorgungskosten bewertet. Aufgrund dieser Vorgehensweise war es möglich, Vorteile und Nachteile des UPSWING Verfahrens zu identifizieren und grundlegend zu beschreiben.

Im Folgenden wird der Inhalt der Arbeit in einer kurzen Übersicht der jeweiligen Kapitelinhalte wiedergegeben:

In **Kapitel 1** wird die Motivation der Arbeit beschrieben. Dazu werden gängige Verfahren zur thermischen Verwertung von Abfällen diskutiert und die Notwendigkeit von neuen, innovativen und effizienten Verfahren vor dem

Hintergrund eines weltweit steigenden Energiebedarfs sowie einer zunehmenden Entsorgungsproblematik herausgearbeitet. Darauf aufbauend werden die Schwerpunkte abgeleitet, an denen der UPSWING Prozess bewertet werden soll.

Kapitel 2 beschreibt den Stand der Technik. Es wird insbesondere auf die konventionelle Müllverbrennung und die direkte Mitverbrennung eingegangen, da der UPSWING Prozess mit beiden vorgenannten Verfahren verglichen werden soll. Des Weiteren wird das Potential des UPSWING Prozesses im europäischen Kontext abgeschätzt.

Kapitel 3 erläutert den experimentellen Ansatz der vorliegenden Arbeit sowie die notwendigen Grundlagen und Randbedingungen. Dabei wird zunächst der UPSWING Prozess detailliert beschrieben und die verfahrenstechnischen Randbedingungen definiert. Von besonderer Bedeutung für den Prozess ist hierbei die partielle Rauchgasreinigung, deren Aufgabe es ist, kritische Rauchgaskomponenten wie Flugstaub, Schwermetalle und Chlor vor Einbringung in den Kraftwerksprozess abzuscheiden. Weitere Komponenten wie Schwefeloxide, Stickoxide und Dioxine werden nicht abgeschieden, da diese im Kraftwerksprozess reduziert bzw. zerstört werden sollen.

Die Überprüfung der Annahmen zur Abscheidung und Schadstoffreduktion ist ein wesentliches Ziel der vorliegenden Arbeit. Dazu werden die unterschiedlichen Aspekte durch einen Vergleich zwischen gekoppeltem Betrieb (UPSWING) und reinem Kohlefeuer untersucht. Ein vergleichbarer Ansatz wird für die direkte Mitverbrennung von Sekundärbrennstoffen gewählt, da beide Verfahren vor dem Hintergrund der hohen ökologischen Standards der konventionellen Müllverbrennung verglichen werden sollen.

In **Kapitel 4** werden die Versuchsanlagen beschrieben, die für die diversen Untersuchungen herangezogen wurden. Notwendige Umbaumaßnahmen und Anpassungen, die sich im Zuge der Untersuchungen ergeben haben, werden angesprochen. Ergänzend werden die im Kontext wichtigen Messverfahren für Quecksilber und Chlor vorgestellt.

Kapitel 5 ist das zentrale Kapitel der vorliegenden Arbeit und fasst die durchgeführten experimentellen Untersuchungen zusammen. Die Ergebnisse werden für die jeweiligen Prozesse diskutiert und verglichen. **Kapitel 5.1** befasst sich mit dem Reduktionspotential für Stickoxide. Es konnte nachgewiesen werden, dass sowohl das UPSWING Verfahren als auch die direkte Mitverbrennung potentiell geringere Emissionen als der reine Kohlebetrieb ermöglichen. Bei der direkten Mitverbrennung ergibt sich dieses Potential durch den höheren Gehalt an flüchtigen Bestandteilen im Vergleich zur Kohle. Beim UPSWING Verfahren wurde festgestellt, dass der Ort der Einbringung der Rauchgase in den Kraftwerksprozess im Hinblick auf die Stöchiometrie im Brennerbereich von entscheidender Bedeutung ist. Eine gute Kontrolle der Prozessbedingungen ist aus verfahrenstechnischer Sicht zwingend erforderlich.

Im Bezug auf Chlor (**Kapitel 5.2**) ergibt sich ein klarer Vorteil des UPSWING Verfahrens. Aufgrund der Abscheidung der Chlorfracht vor dem Einbringen der Rauchgase in den Kraftwerksprozess kommt es zu einer Verdünnung der gemischten Rauchgase. Weiterhin wird keine abfallstämmige Asche und damit kein aschegebundenes Chlor oder Alkalien in den Kraftwerksprozess eingebracht. Im Falle der direkten Mitverbrennung erhöht sich die Konzentration von gasförmigem Chlorwasserstoff in den Rauchgasen. Dieser Aspekt alleine lässt jedoch noch nicht auf mögliche Betriebsbeeinträchtigungen schließen. Allerdings wurde bei den Versuchen mit Braunkohle eine deutliche Erhöhung des aschegebundenen Chloranteils festgestellt, was auf mögliche Betriebsprobleme durch Korrosion schließen lässt, deren detaillierte Untersuchung jedoch nicht Gegenstand dieser Arbeit ist.

In **Kapitel 5.3** wird das Zerstörungspotential für Dioxine bzw. Furane (PCDD/F) an zwei Versuchsanlagen untersucht (Staubfeuerung und Wirbelschicht). Dazu wurde im Vorfeld der Versuche Kohle mit Dioxinen beladen, um einen entsprechenden Schadstoffeintrag in den Prozess zu simulieren. Dabei konnte, bei einem konservativen Ansatz, ohne Berücksichtigung einer möglichen DeNovo Synthese, eine Zerstörungseffizienz größer 99,9% festgestellt werden. Wesentliche Unterschiede zwischen den Referenzversuchen und den eigentlichen Versuchen mit Dioxineintrag konnten nicht festgestellt werden. Im Hinblick auf die direkte Mitverbrennung von Sekundärbrennstoffen stützen sich die Aussagen auf Ergebnisse eines Großversuchs an einem deutschen Braunkohlekraftwerk. Die im Rahmen von Abnahmemessungen gemäß 17.BImSchV durchgeführten Untersuchungen ergaben keine erhöhten Dioxinemissionen.

Kapitel 5.4 befasst sich mit Schwermetallen, insbesondere mit Quecksilber. Dabei wurde im Hinblick auf das UPSWING Verfahren der Einfluss der in den Kraftwerksprozess eingebrachten Rauchgase auf das Verhalten und die Verteilung der Schwermetalle untersucht. Dabei konnte ein deutlicher Einfluss des Wassergehaltes der Rauchgase auf die Speziation von Quecksilber nachgewiesen werden. Ähnliches gilt für Chlor, das aber durch entsprechende Verfahren vor dem Einbringen der Rauchgase in den Kraftwerksprozess zu einem großen Teil abgeschieden wird. Im Gegensatz hierzu wird bei der direkten Mitverbrennung eine zusätzliche Fracht in den Kraftwerksprozess eingebracht. Bei den Elementen Zink, Blei und Kupfer wurden deutlich erhöhte Konzentrationen in der Flugasche gemessen. Dagegen konnten für Quecksilber nach Aschefilter keine höheren Emissionen festgestellt werden.

Kapitel 5.5 befasst sich mit der Flugaschequalität. Dieser Aspekt wurde hauptsächlich für die direkte Mitverbrennung untersucht, da im UPSWING Verfahren keine Asche oder Flugstaub in den Kraftwerksprozess eingebracht wird. Grundlage der Untersuchungen sind die Anforderungen an die Flugasche gemäß DIN/EN 450. Im Rahmen dieser Versuche konnte insbesondere für Chlor und Schwefel ein deutlicher Einfluss der Mitverbrennung auf die

Aschezusammensetzung festgestellt werden. Ein wesentliches Ergebnis dieser Untersuchungen ist die Erkenntnis, dass bei einer Beurteilung der Auswirkungen auf die Flugaschequalität immer das spezifische System Sekundärbrennstoff – Hauptbrennstoff betrachtet werden muss.

In Ergänzung zu den experimentellen Untersuchungen in Kapitel 5 wird in **Kapitel 6** die großtechnische Umsetzung des UPSWING Verfahrens mittels CFD Simulation untersucht. Dabei geht es primär um die Auswirkung der Rauchgaseinbindung in den Kraftwerksprozess sowie um die Beurteilung unterschiedlicher verfahrenstechnischer Varianten zur Eindüsung. Für den untersuchten Braunkohlekessel konnte die Eindüsung über die Brüdenbrenner als beste technische Lösung identifiziert werden. Als Alternative bietet sich ebenfalls die Eindüsung über den Aschetrichter an. Generell wurde allerdings festgestellt, dass die Eindüsung einen negativen Einfluss auf den Kraftwerksprozess hat, da aufgrund der höheren Abgasmenge die Abgasverluste ansteigen und der Wirkungsgrad sinkt. Durch eine entsprechende Begrenzung der eingebrachten Rauchgasmenge lassen sich diese Verluste minimieren. Für den untersuchten Braunkohlekessel wurde eine anteilige Rauchgasmenge von ca. 5% bezogen auf die Gesamtrauchgasmenge als sinnvolle Begrenzung identifiziert.

Kapitel 7 fasst die Ergebnisse der Arbeit zusammen. Daneben werden weitere prozessrelevante Aspekte diskutiert, die im Rahmen der experimentellen Untersuchung nicht oder nicht abschließend beurteilt werden konnten. Die Ergebnisse der Arbeit können wie folgt zusammengefasst werden:

Aus umwelttechnischer Sicht genügt das UPSWING Verfahren den hohen Standards einer konventionellen Müllverbrennung. Im Hinblick auf Schadstoffe wie Stickoxide oder Dioxine, die in den Kraftwerksprozess eingebracht werden, konnte eine sichere Reduzierung bzw. Zerstörung festgestellt werden. Hingegen muss im Hinblick auf die direkte Mitverbrennung von einem Schadstoffeintrag in den Kraftwerksprozess ausgegangen werden. Speziell bei Schwermetallen hängt dieser Eintrag stark von der Qualität der Brennstoffe bzw. der Ausgangsfraktionen ab. Im Falle von modernen Kraftwerksanlagen kann auch hier von einer sicheren Abscheidung der Schadstoffe ausgegangen werden, beispielsweise über die Flugasche oder REA Produkte. Eine Verunreinigung der Kraftwerksnebenprodukte muss allerdings akzeptiert werden. Vor dem Hintergrund einer nachhaltigen und sicheren Schadstoffkontrolle liegen die Vorteile damit auf Seiten des UPSWING Verfahrens.

Im Gegensatz zeigt der Vergleich der verfahrenstechnischen Umsetzung beider Prozesse Vorteile für die direkte Mitverbrennung. Dies gilt insbesondere hinsichtlich der Prozessintegration in einen bestehenden Kraftwerksprozess, der Flexibilität, beispielsweise bei Lastwechsel, und bei Notfallsituationen. Generell erscheint die Verfahrenstechnik für den gekoppelten UPSWING Prozess schwieriger. Zwei Anlagen müssen gemeinsam betrieben werden, was die

Flexibilität in jedem Fall einschränkt. Weiterhin erhöht sich der gesamte leit- und regelungstechnische Aufwand. Betriebsprobleme einer Anlage werden immer Auswirkungen auf die jeweilig andere Anlage haben. Aufgrund dieser Thematik wird deshalb die Kopplung einer Rostfeuerung an mindestens zwei Kohlekessel empfohlen. Aus wärmetechnischer Sicht darf auch der Einfluss der feuchten Rauchgase nicht unterschätzt werden. Eine Verschiebung des ausgekoppelten Wärmestroms in die Nachschaltheizflächen erscheint wahrscheinlich. Im Vergleich dazu ergeben sich für die direkte Mitverbrennung weitaus weniger verfahrenstechnische Probleme bei der Umsetzung. Eine separate Dosierung erlaubt eine gezielte Einbringung des zusätzlichen Brennstoffes in den Kessel während des Normalbetriebs. In kritischen Betriebsphasen wie beispielsweise Anfahr- und Abfahrvorgängen kann auf den zusätzlichen Brennstoff verzichtet werden. Demzufolge verspricht die Mitverbrennung weitaus weniger Probleme bei der Umsetzung an einer bestehenden oder neu zu planenden Anlage. Einzige Ausnahme hiervon ist der erhöhte Chloreintrag, der vor dem Hintergrund möglicher Korrosionserscheinung sowie Kontamination von Kraftwerksnebenprodukten (Flugasche, REA-Produkte) von Fall zu Fall untersucht werden muss.

Im Hinblick auf die ökonomische Seite zeigen beide Prozesse ein deutliches Potential die direkten Kosten für die thermische Verwertung von Abfällen zu reduzieren. Beim UPSWING Konzept liegt der Vorteil auf der Investitionsseite, da bei wesentlichen Komponenten wie Turbine/Generator bzw. der Rauchgasreinigung auf die im Kraftwerksprozess bereits vorhandenen Komponenten zurückgegriffen werden kann. Ferner ist ein Vorteil, dass der Abfall ohne weitere Aufbereitung in der Rostfeuerung eingesetzt werden kann. Bei der direkten Mitverbrennung sind am Kraftwerksstandort nur geringe Investitionen für Lagerung, Transport und ggf. einer zusätzlichen Aufbereitung erforderlich, da die wesentlichen Kosten im Zuge der Herstellung der Sekundärbrennstoffe anfallen und diese in der Regel nicht vom Kraftwerksbetreiber zu tragen sind.

Zusammengefasst muss die Frage gestellt werden, ob der zusätzliche Aufwand für das UPSWING Verfahren gerechtfertigt ist. Aus ökologischer Sicht muss diese Frage mit ja beantwortet werden, insbesondere vor dem Hintergrund einer nachhaltigen und sicheren Schadstoffkontrolle. Aufbauend auf den durchgeführten Untersuchungen wäre der nächste Schritt konsequenterweise die großtechnische Demonstration des Verfahrens.

1 Introduction

1.1 State-of-the-art waste disposal concepts

In most European Countries waste disposal basically relies on landfill. However, with the implementation of the European Waste Landfill Directive the disposal of untreated waste is or will be omitted in the near future. With the implementation in June 2005, some European countries ran into significant capacity problems. Germany is one example, as waste treatment capacities are still insufficient. As a result, waste has to be temporarily stored. Other European countries still have some time to develop and improve their current waste management systems. Nevertheless the lack of environmental-friendly disposal capacities is and will be an enormous challenge in the next decade.

In most developed countries, incineration, with energy recovery in the form of electricity and/or useful heat, and the utilisation/landfill disposal of the solid residues, is one of the principal elements of integrated management systems for municipal solid waste (MSW). Commonly used are grate firing systems with capacities ranging from 50,000 up to 500,000 Mg/a. Some European countries, such as Germany and the Netherlands, have significant incineration capacities installed. In Germany, the available incineration capacity was approx. 18 Mio. Mg/a in the year 2006 (see Fig. 1-1) including facilities currently under construction.

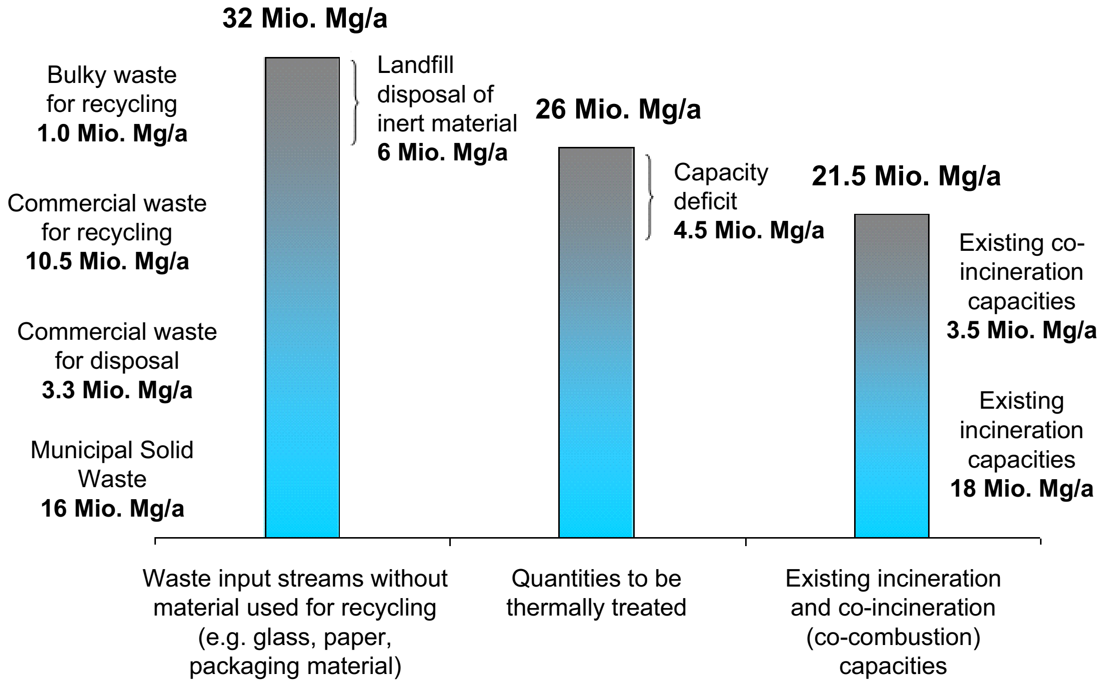

Fig. 1-1: Waste management scenario (Germany) and quantities for the year 2006 [1]

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[1] PROGNOS AG, VGB PowerTech, 10/04, (2004)

In addition to incineration, approx. 3.5 Mio. Mg/a of waste are currently co-incinerated, primarily in industrial furnaces such as lime- or cement kilns, but also in industrial processes like smelters, where they partly replace valuable fossil resources. In order to prevent misunderstandings, these waste materials do not consist of untreated MSW, but of selected mono fractions with considerable energy content. Taking incineration and co-incineration capacities into account, approx. 21.5 Mio. Mg/a can be thermally treated. Nevertheless the total waste input stream does not consist of MSW only. Considering all relevant waste streams (including commercial waste) without materials used for recycling up to 26 Mio. Mg/a have to be thermally treated, thus resulting in a capacity deficit of some 4 to 5 Mio. Mg/a in the year 2006 [1].

Incineration processes, however, have been subject to increased capital and operating costs due to the improvements - required by legislation - in their environmental performance. To overcome missing capacities and economic deficits other technologies are sought. As already mentioned, co-incineration is a technically feasible and short-term available solution, which is commonly applied in industrial processes. The co-incineration of pre-treated waste materials in thermal power plants is a further step - offering high-efficient waste-to-energy conversion - and could be considered as an emerging market in the future.

In this context, the production of fuels from waste materials suitable for utilisation in thermal power plants is a challenging development. Contrary to industrial processes like kilns or smelters, the technical demand upon such waste-derived fuels is considerably higher. With the availability of new and reliable automatic sorting technologies such as near-infrared detection and separation (NIR), their production has become technically feasible and economically attractive [2]. The idea behind this concept is to pick valuable materials such as mixed plastics from the raw waste stream and convert them into a combustible form. Therefore, the production process includes further mechanical process steps such as crushing, classifying, drying and ferrous/non-ferrous separation. The long-term operational effects and environmental compatibility of such waste-derived fuels - in the following referred to as Solid Recovered Fuels (SRF) - are currently under development and demonstration.

Going beyond conventional incineration or co-incineration, advanced concepts refer to different technological approaches such as gasification and pyrolysis processes, or the combination of different process steps, e.g. gasification followed by combustion. Despite of the projected benefits and advantages of such technologies, the results achieved in industrial scale were often negative and economical unattractive [3].

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[2] Th. GLORIUS: „Erfahrung mit Produktion und Einsatz gütegesicherter Sekundärbrennstoffe – RECOFUEL“, 11.Fachtagung Thermische Abfallbehandlung, München, 14-15.03.2006, ISBN-10: 3-89958-198-9, (2006)

[3] Siemens/KWU: „Keine Akquise in Deutschland“, Entsorga-Magazin 5, S. 121, (1999)

The combination of separate facilities is a further approach to provide economical and environmental solutions in the future. An example for an innovative combined process is the so-called UPSWING process. UPSWING is the acronym for Unification of Power Plant and Solid Waste INcineration on the Grate, describing the combination of a conventional grate firing system with a power plant both on the steam- and the flue gas side. The concept was developed by the Forschungszentrum Karlsruhe, Germany, and patented 1998-2003 [4]. However, it has to be emphasised that the UPSWING process is not realised up to now, neither in demo- nor full-scale application.

The UPSWING concept was recent subject of the European Research Project UPSWING (ENK5-CT-2002-00697), which was successfully finished in 2006. Based on the results of this project, the present thesis focuses on this new and promising process, its environmental, economical, and operational advantages being compared with other available technologies such as conventional waste incineration and co-incineration.

The following descriptive part will give a more-detailed insight of the relevant processes, further discusses the demand for alternative waste disposal concepts, and concludes with the methodology used in this thesis.

1.1.1 Municipal Solid Waste Incineration (MSWI)

The typical system for MSWI in Central Europe is the so-called European mass burner, a facility which incinerates the waste on the grate without prior treatment. Often, waste incineration is referred to as "mono-combustion", although the term "incineration" fits better to the grate firing concept. Anyway, the chemical energy of the waste is released as heat in the combustion process and transferred to the boiler system. Modern boilers recover more than 80% of this heat. The steam can be utilised in different ways such as district heating purposes, production of electrical power, direct utilisation in industrial processes, or a combination of the mentioned utilisation paths.

MSWI has relatively low electricity generation efficiencies, resulting from the poor quality of MSW as a boiler fuel and the relatively low steam parameters achievable, due to concerns about excessive corrosion rates of high temperature boiler components. In case of power generation, modern plants reach an average electrical efficiency of 18%, while modern plants can reach up to 25%. Optimum efficiency is achieved by a combination of power and heat utilisation (combined heat and power, CHP). Using CHP, energy recovery in the range of 70 to 75% becomes possible [5].

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[4] H. HUNSINGER, S. KREISZ, H. SEIFERT, J. VEHLOW: "Verfahren zur Beschickung der Verbrennungseinheit eines Kohlekraftwerks"; DP-OS 19 723 145 (10.12.1998), DP-PS 19 723 145 (8.8.2002), EP-PS 59 804 147 (15.5.2002), JP-PS 3 392 424 (24.1.2003), IL-PS 132 336 (18.12.2003); (1998, 2002, 2003)

[5] E. DIRKS: „Praxishandbuch Abfallverbrennung – Technik und Betrieb thermischer Behandlungsverfahren", Herrentor Fachbuchverlag, ISBN 3-00-005535-5, (2000)

1.1.2 Co-combustion of Solid Recovered Fuels (SRF)

Co-incineration (or co-combustion) of pre-treated waste-derived fuels such as SRF describes their co-utilisation in industrial furnaces or power plants as a supplement fuel. The intention is to replace a certain amount of the regular fossil fuel (coal, oil, gas), mainly because of economic reasons. In respect to the closed combustion system of a power plant, where coal is combusted and not incinerated, the term “co-combustion” will be applied for SRF, accordingly.

Complying with the emissions and air quality control directives, co-combustion of SRF can be an efficient and low-cost form of energetic and material exploitation. The high biomass content of SRF (usually > 50%) is an additional means to use a substantial potential in a highly-efficient and cost-effective way in power generation, thus making a lasting contribution to CO_2 emission reduction and resource saving. Nevertheless, co-combustion processes do not provide secure control of a number of the pollutant species present in the waste stream, and particularly of the heavy metals and the halogens. Hence, the investigation of this potential disadvantage becomes one of the primary objectives of this thesis.

1.1.3 Combined processes

An early concept realising this principle was the so-called “Satellite Combustion”, developed by the German Company “Hölter” together with the former German Engineering Company “Steinmüller”. Hot flue gases of a waste incinerator were directly transferred into the boiler of a power plant. The main problem of this concept is that all pollutants are transferred to the power plant process. Disturbance of the power plant process, e.g. by exceeding emission limits, increased corrosion problems (potentially induced by chlorine), or deterioration of power plant residues, could be expected.

The UPSWING process can be considered as a subsequent improvement of the Hölter Process. The UPSWING process includes partial flue gas cleaning of the waste flue gases prior to injection into the coal boiler. As a consequence, the risk of negative effects on the power plant process should be omitted. Furthermore, the steam produced in the boiler system of the waste incinerator is included in the steam circuit of the power plant. As a result, increased efficiency of waste-to-energy conversion can be expected. As the focal subject of this thesis, the process and its boundary conditions will be discussed in chapter 1.3.

1.2 The demand for alternative waste disposal concepts

The European Landfill Directive (1999/31/EC) sets strict rules on landfill disposal of untreated MSW in the EU countries and forces a reduction of the biodegradable quantities disposed off to landfills up to 35% of the amount produced in 1995 in the coming decade. More environmentally friendly waste management options are promoted under the framework of the European Waste Strategy (1996), which lays down the hierarchy of waste management policy as follows: (1) Prevention of waste; (2) Recovery (material over energy); and (3) Final disposal.

This hierarchy must be applied with certain flexibility and be guided by considering the best environmental solution taking into account economic necessities and social requirements. Where environmentally sound preference should be given to material over energy recovery, although in certain cases preference can be given to energy recovery. Considering the application of a preferable option a clear definition is still required in most member states.

Therefore, it is necessary to point out the environmental benefits of thermal waste treatment with heat- and energy recovery. The concept comprises not only the protection of human health and environment, but is also capable to conserve fossil fuels by energetic utilisation of residues and waste. This aspect leads to an issue with increasing importance: Energy recovery from waste and residues can significantly contribute to climate protection by avoidance of organic emissions from landfills (e.g. methane) and reduced CO_2 emissions by replacing fossil fuels. Furthermore, approx. 50% of MSW can be attributed to as biomass, leading to an additional benefit in terms of CO_2 emission reduction. The utilisation of waste is therefore fully complying with the requirements of the European Directive 2001/77/EC on the promotion of electricity from renewable sources.

Considering the global development of population and energy consumption (see Fig. 1-2), the future importance of energetic utilisation of waste becomes obvious. Despite of all efforts to safe energy and to improve efficiency, worldwide energy consumption will increase dramatically, especially in developing and emerging countries.

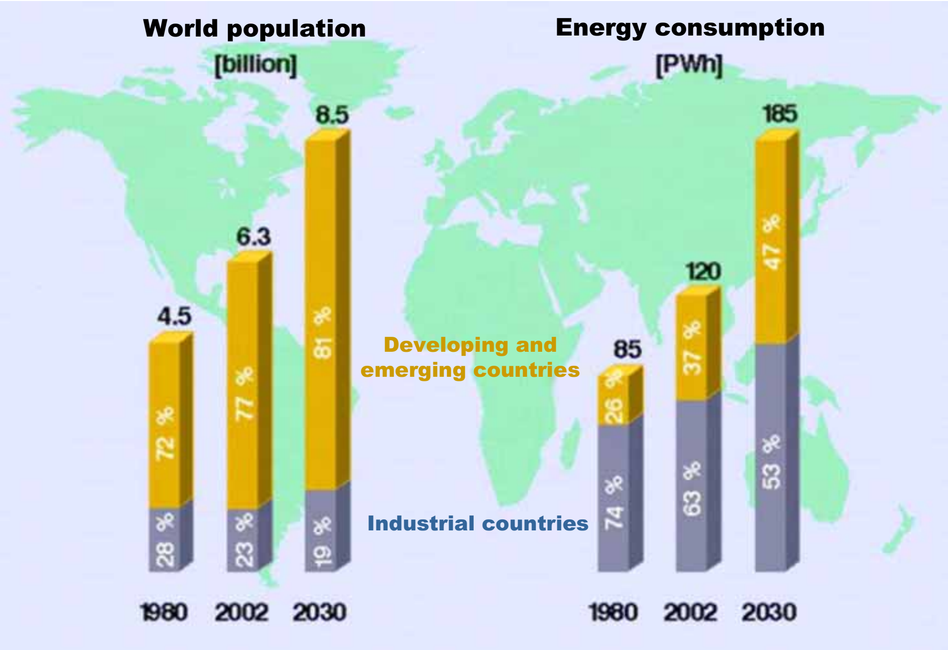

Fig. 1-2: Forecast of world population and energy consumption [6]

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[6] International Energy Agency (IEA), Forecast of world population and world energy consumption, (2002)

However, the disproportion between developing and industrial countries will remain. For the time being 25% of the world population consume approx. two-thirds of the total energy supply. The utilisation of waste for heat- and electricity production can significantly contribute to the reduction of necessary energy imports. In this context more efficient and sustainable waste treatment policies and technologies become gradually necessary within the EU and worldwide. In view of these demands, the UPSWING process was developed as an advanced waste treatment concept.

1.3 UPSWING as an advanced waste treatment concept

The UPSWING process, describing the integration of a waste incinerator on the flue gas and steam sides to a large coal-fired boiler, has been developed to overcome the economical deficits of conventional MSWI while maintaining its environmental advantages [7][8]. A schematic overview of the UPSWING process is given in Fig. 1-3, covering the waste-to-energy section, the partial flue gas cleaning concept, as well as the integration of both steam- and waste flue gas to the power plant.

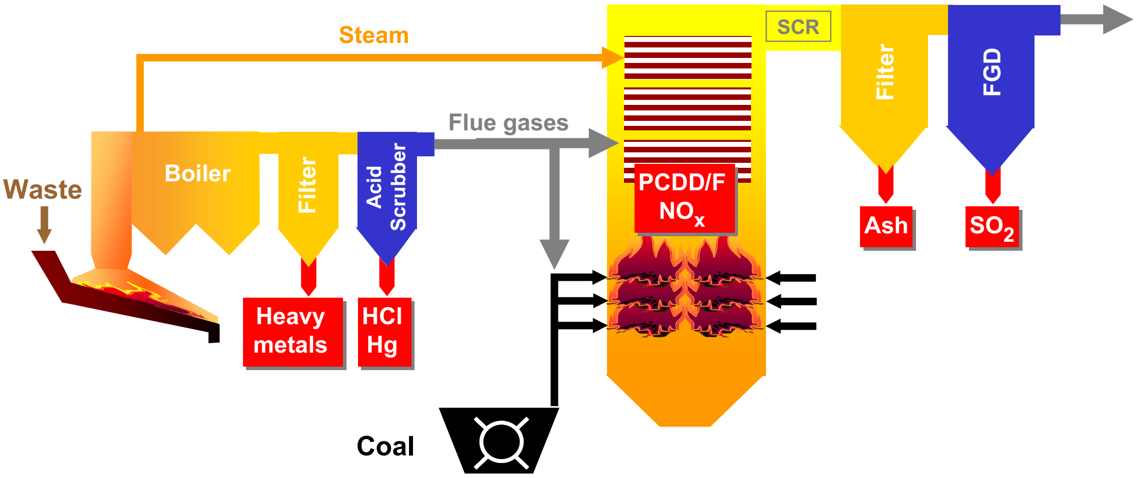

Fig. 1-3: The UPSWING concept

The waste-to-energy section consists of a grate firing and boiler system comparable to those utilised in modern MSWI. This is an important aspect, as reliable technology is available on the market, which can be used without the need for further research- or technological development. The steam produced by the waste is forwarded to the boiler system of the connected power plant. Hence, increased steam parameters can be achieved and its expansion in the generator unit of the power plant leads to higher electrical efficiency. Furthermore, the waste flue gases - partially cleaned - are injected into the combustion chamber.

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[7] J. VEHLOW, H. HUNSINGER, S. KREISZ, H. SEIFERT: „UPSWING - Kombination von Abfallverbrennung und Kohlekraftwerk“, In: Schriftenreihe des Fachgebietes Abfalltechnik Universität Kassel (Hrsg.: Urban, A.I.), 67 – 82, Kassel, Germany, (2003)

[8] J. VEHLOW, H. HUNSINGER, S. KREISZ, H. SEIFERT: “UPSWING - A novel concept to reduce costs without changing the environmental standards of waste combustion”, IEA Bioenergy Joint Task Seminar, Tokyo, October 28, (2003)

1.3.1 Flue gas integration and partial flue gas cleaning

The idea behind the partial flue gas cleaning concept is to keep critical substances away from the power plant process, thus limiting environmental and operational disadvantages. The partial flue gas treatment system is based on de-dusting of the waste flue gases using conventional bag-filter systems (BFS) or electrostatic precipitators (ESP). The major part of critical heavy metals is removed with the fly ash. Acid components like HCl, HBr and HF, imposing significant risks for boiler operation, are removed in an acid scrubber, which is part of the partial waste flue gas treatment process. The partially cleaned gas, which still contains SO_2 and NO_x as well as gaseous PCDD/F and other organic pollutants, is introduced to the power plant process. The remaining gaseous PCDD/F is totally destroyed inside the combustor. NO_x and SO_2 will be reduced to acceptable levels by the air pollution control system (APC) of the boiler.

1.3.2 Steam integration

Steam integration refers to the utilisation of the waste energy in the turbine/generator set of another plant (e.g. combined cycle turbine). A comparable approach was realised in Mainz, Germany, by company Martin GmbH. Steam produced from MSWI (40bar and 400°C) is connected with a combined cycle natural gas turbine as shown in Fig. 1-4.

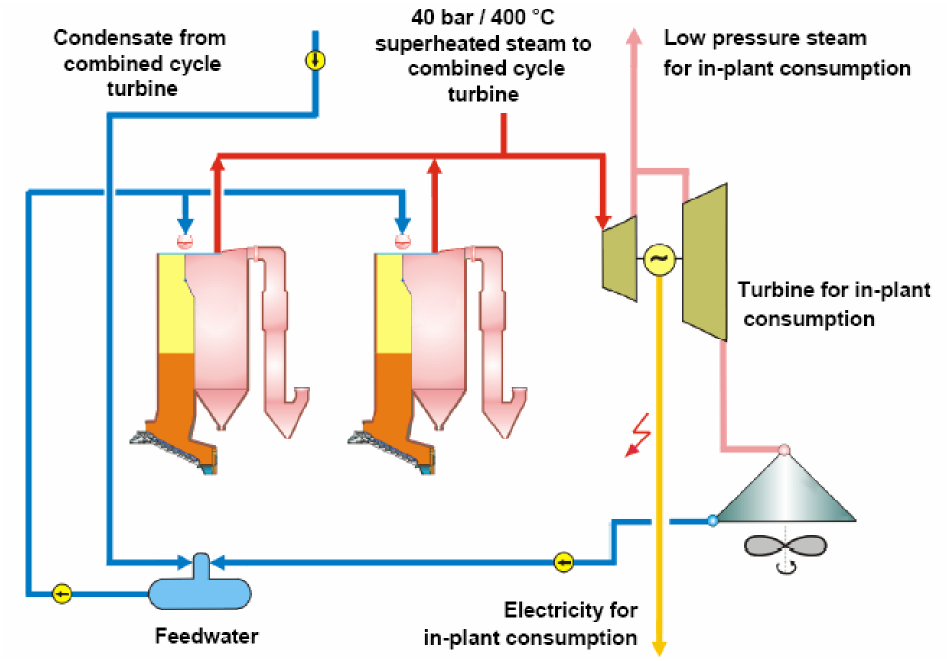

Fig. 1-4: Combination of MSWI and combined cycle gas turbine, Mainz, Germany [9]

Steam produced from MSWI (40bar and 400°C) is connected with a combined cycle natural gas turbine. The superheated steam produced is brought forward to a combined-cycle gas turbine process, where it is overheated and expanded in the combined cycle steam turbine. The condensate is returned to the MSWI feedwater tank. The MSWI plant is still equipped with a medium and low

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[9] J. MARTIN: "Global use and future prospects of waste-to-energy technologies", Waste-to-Energy Research and Technological Council, Columbia University (USA), October 7-8, (2004)

pressure turbine in order to guarantee high operational flexibility. The combined electrical efficiency of this combined process was determined to be higher than 40%, which is far beyond conventional MSWI. A comparable concept was realised in Bilbao, Spain. Both examples show that steam integration is generally possible, although the efforts for integration to a coal-fired power plant in case of the UPSWING concept are expected to be significantly higher.

1.3.3 Expected benefits

The UPSWING concept basically relies on state-of-the-art technology; a development of new components is not required. Combination of the steam circuits of both facilities proposes higher net electrical efficiency in waste-to-energy conversion, leading to lower CO_2 emissions in electricity production. Furthermore, a reduction of the specific investment costs in comparison to a same-sized standalone MSWI can be expected. All factors should result in a lower gate fee per Mg (or tonne) of waste to be disposed of. In comparison to alternative waste-to-energy concepts, the UPSWING concept proposes the same stringent environmental standards as conventional MSWI. One of the major benefits of the UPSWING concept is the integration to existing power plants. Approx. 23.5% of the world energy consumption and 38% of the world energy production are covered by the utilisation of coal [10]. With this background the concept is especially favourable for those countries which largely rely on fossil fuels and lack of state-of-the-art waste treatment facilities, e.g. Poland, Romania, and Bulgaria.

1.4 Methodology

1.4.1 Problem definition and primary objective

The UPSWING concept is proposed as an advanced technical solution to solve economical deficits of conventional thermal waste treatment as well as environmental/operational deficits of alternative waste treatment concepts such as direct co-combustion. However, the UPSWING process is still a (theoretical) concept and not realised yet. A direct comparison is therefore not possible. Nevertheless, a method was sought which should allow exactly such a process comparison, and to qualify the UPSWING process as a potential option in a future thermal waste treatment scenario. This approach is discussed in chapter 1.4.2, with the initial idea to compare both UPWING and direct co-combustion and subsequently elaborate the advantages respectively disadvantages of the one or the other concept.

1.4.2 Approach and areas of concern

The question is now how to approach to such an ambitious task. Taking a look on the proposed benefits of the UPSWING concept, already discussed in the previous chapters and summarised in Fig. 1-5, may help to identify the relevant aspects, worth to be taken into consideration:

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[10] Gesamtverband des deutschen Steinkohlebergbaus, Steinkohle Jahresbericht, (2003)

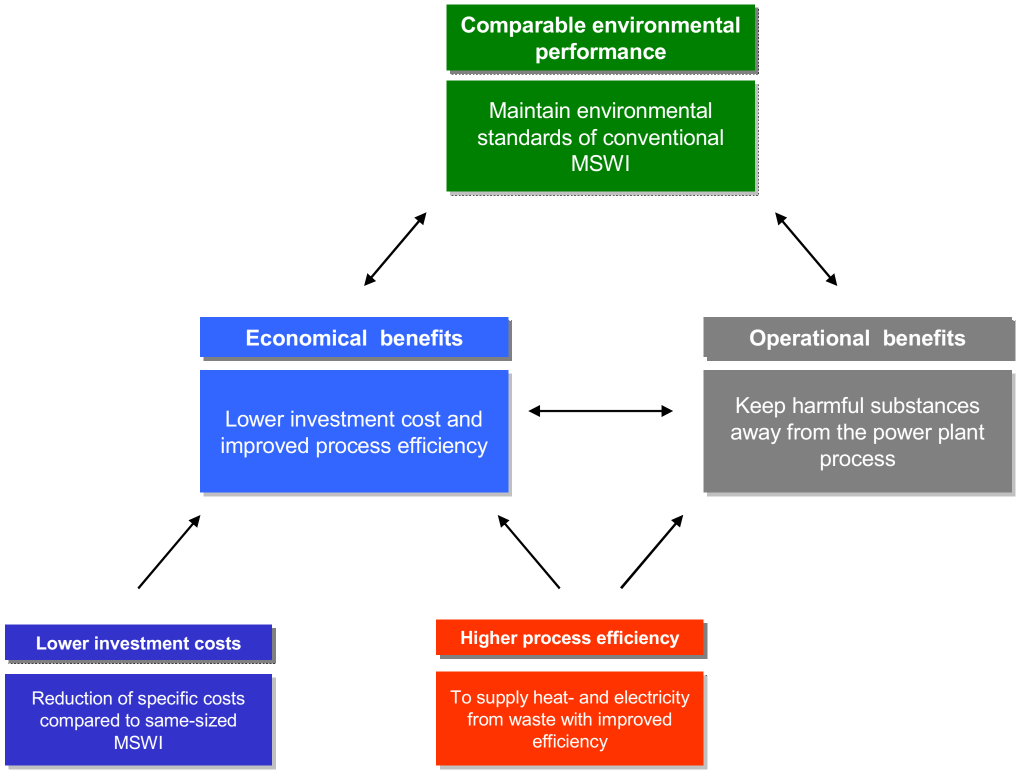

Fig. 1-5: UPSWING: Environmental, economical (efficiency/investment), and operational benefits

The diagram shows three areas of concern, comprising of environmental, operational, and economic benefits. From an economical point of view, UPSWING proposes lower investment costs and improved process efficiency (waste-to-energy) in comparison to conventional MSWI. In comparison to direct co-combustion of waste-derived fuels such as SRF in conventional power plants, UPSWING proposes to maintain environmental standards of MSWI. Furthermore, potential harmful substances are not brought forward into the power plant process, as it is the case for direct co-combustion.

Coming back to the initial idea, which is the qualification of the UPSWING process as a potential option in a future thermal waste treatment scenario, leads to the following approach:

- Investigation and evaluation of the performance of the UPSWING process in the relevant areas of concern,
- Investigation and evaluation of the performance of alternative concepts, with the focus on direct co-combustion, in the same areas of concern,
- Process comparison and assessment of the potential of the UPSWING concept

From a technical point of view, and due to the fact that any new concept has to compete on the market, the mentioned areas are considered crucial for a successful realisation of the UPSWING concept, and, furthermore adequate to evaluate the process. Therefore, a closer look on these areas appears to be necessary:

(a) Environmental aspects

An important aspect is the evaluation of the environmental compatibility of the UPSWING process. The process emissions have to be investigated and evaluated against the standalone power plant process, but also against standalone MSWI. As the main experimental part of this work, direct process emissions, destruction potential of PCDD/F, the fate of the toxic element mercury, as well as fly ash quality and trace element behaviour were investigated. Following the discussed approach, the UPSWING concept has to prove its environmental comparability to mono-combustion (MSWI). Consequently, destruction of PCDD/F and reduction of pollutants not influenced by the partial flue gas cleaning (NO_x, SO_2) are of major concern. Furthermore, the influence of SRF co-combustion on process emission and residue quality was investigated in detail in order to evaluate the potential benefits of the partial flue gas cleaning concept, and to compare both processes, accordingly.

(b) Operational aspects

The impact of the flue gas integration on the power plant process is an important aspect of the UPSWING process to be examined, as unrestricted boiler operation is - from the view of the power plant operator - crucial for successful implementation. The impact of flue gas integration on the power plant process was primarily investigated by full-scale boiler simulation. Additional aspects related to control/regulation of two separate facilities were considered as well. The consequences of flue gas integration were compared with those of direct SRF co-combustion, whereas the element chlorine was selected as the leading component.

(c) Economical aspects

Both process efficiency (waste-to-energy) and investment cost are related to financial aspects and can be summarised as “economical aspects”, accordingly. Considering economical aspects, an important parameter is the so-called “gate-fee”, the actual- or estimated costs for waste disposal, usually expressed in €/Mg waste, because - despite all potential environmental benefits - all new and advanced waste treatment concepts obviously have to compete on the market. Nevertheless a comparison to landfill disposal is not fair and - following the European Landfill Directive - landfill disposal of untreated MSW is no further option. Therefore the actual gate-fees of municipal solid waste incineration are considered as reference values (100%). In order to quantify potential economical benefits of the UPSWING concept, a theoretical gate-fee was calculated and compared with the gate-fee of a standalone MSWI of the same size.

2 State of knowledge

The previous chapter focuses on the UPSWING process as a new and promising waste treatment concept. The need for such concepts was discussed behind the background of the urging demand for alternative waste disposal concepts. Other state-of-the-art waste disposal concepts such as MSWI or direct SRF co-combustion were briefly introduced. Due to its importance for the UPSWING process, the following chapter will give a more detailed description of modern MSWI, as major parts of the waste-to-energy section and flue gas treatment system are comparable. Furthermore, SRF production and utilisation as an alternative waste treatment concept has to be considered as well, as this information is required to compare both concepts, accordingly. Chapter 2.3 focuses on the current situation on the European waste- and power market. Finally, relevant environmental regulations, applicable for both processes, will be discussed in chapter 2.4.

2.1 Modern Municipal Solid Waste Incineration (MSWI)

The waste-to-energy section of the UPSWING process consists of a grate firing and boiler system comparable to those utilised in conventional MSWI. It is therefore necessary to take a look on waste incineration facilities. An example for a modern MSWI is given in Fig. 2-1:

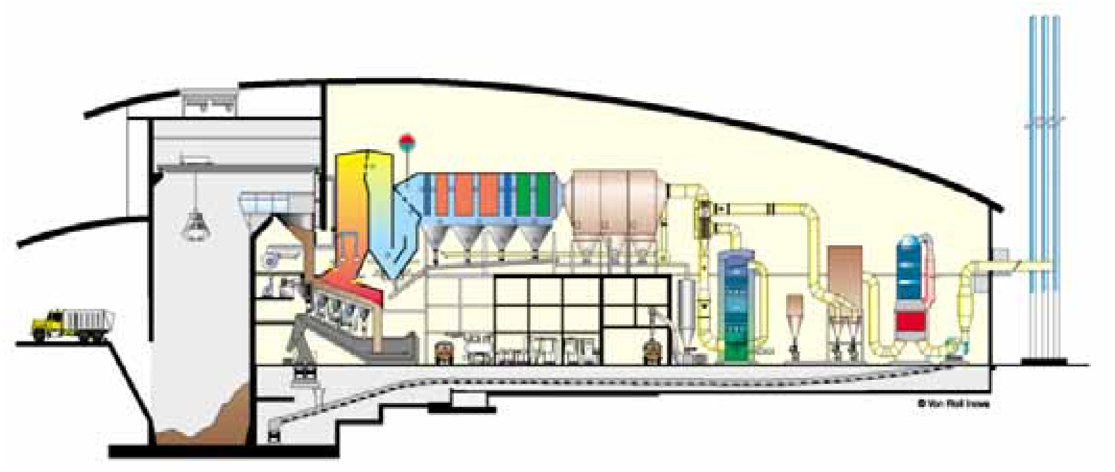

Fig. 2-1: A modern municipal waste incineration (MSWI) plant [Source: VON ROLL INOVA]

The scheme gives a schematic overview of the main process compounds. Waste delivered is stored and separated in the waste bunker. From there, it is fed to the main combustion system, consisting of the grate firing and adjacent boiler system for heat recovery. The heat is commonly utilised in a turbine/generator unit for power production. Following the flue gas behind the boiler, the flue gas cleaning system reduces the concentration of pollutants, evolved during waste combustion, below the required limits. A further import section is the residue treatment, dealing with slag, fly ash, and process water.

2.1.1 Waste reception and storage

The main function of the waste bunker is reception area and temporary storage of the delivered waste. The waste bunker should be large enough to cope with heavy fluctuation in delivery. Furthermore, its capacity should be sufficient to bridge planned and unplanned standstills (usually 10 days maximum before the biodegradable part of the waste starts to decompose). Specially designed bunker cranes use claws to relocate the waste in the bunker, creating a more homogeneous composition, and to transport the waste to a funnel leading to the dispenser. The dispenser is located at the bottom of the funnel and places waste material on the grates using a hydraulic ram or a belt.

2.1.2 Grate firing system, boiler and power production

The grate firing system is the main part of a MSWI plant. The main functions of the grate are carrying and transporting the fuel bed to secure a uniform distribution of the fuel, to supply and distribute primary combustion air, to mix primary air and fuel, and to control fuel bed height and residence time on the grate [11]. Grates are cooled with air, water, or oil. During the incineration process, the waste passes roughly three zones in order to achieve complete combustion. The three zones are the drying/pyrolysis zone, the main combustion zone including gasification with volatile and fixed carbon combustion, and the final burnout zone, as indicated in Fig. 2-2:

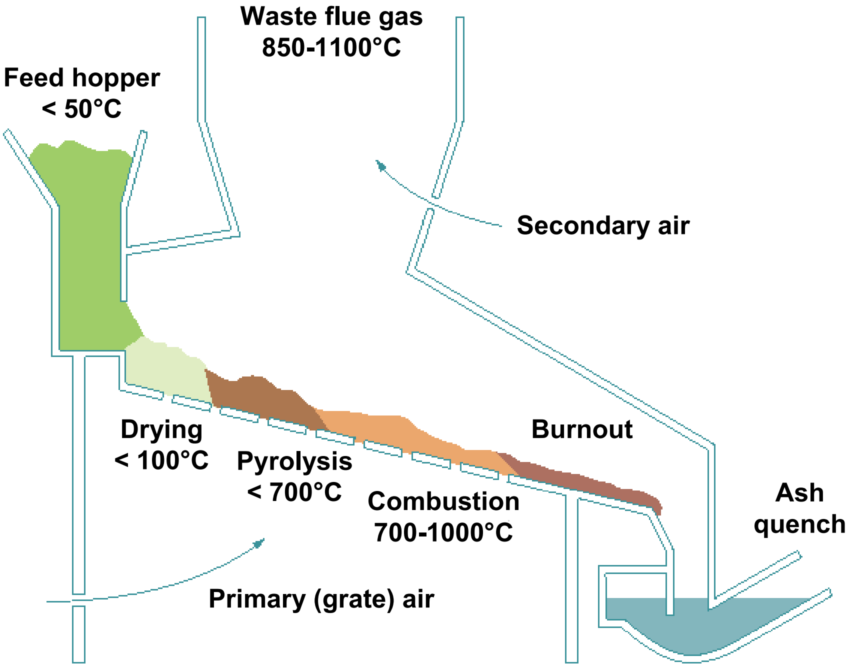

Fig. 2-2: Combustion zones on a typical grate firing system

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[11] R. SCHOLZ, M. BECKMANN, F. SCHULENBURG: „Möglichkeiten der Verbrennungsführung bei Restmüll in Rostfeuerungsanlagen“, BWK Spezial „Müllverbrennung und Entsorgung“, 10/91, S.V22-V39, (1991)

At the end of the grate, the residue and ashes are deposited to a deslagger (ash quench). Secondary air is injected above the fuel bed to achieve final burnout of organic components such as carbon monoxide in the gas phase. Typical grate designs are forward and reverse acting grates, reciprocating grates, and counter-reciprocating grates, whereas the grate elements consist of bars [12]. Other grate designs are roller and travelling grates.

The function of the boiler, located above the grate, is to transfer the heat produced to a medium that makes further utilisation possible. This medium is mostly steam or hot/warm water. If the heat is used for (domestic) heating, hot/warm water boilers and low-pressure steam boilers can be used. If the heat is used for electricity production or for process steam to the industry, the aim would be higher steam parameters (steam temperature and pressure). The boiler consists of two parts, a radiation and a convection part. In the radiation part, the flue gases from the furnace are cooled from about 1100°C to about 600°C. In the convection part, the flue gases are further cooled to about 200°C. Steam from the boiler can be directed to a turbine where it expands. The turbine is connected to a generator where the mechanical energy is converted to electrical energy.

2.1.3 Flue gas cleaning system

The flue gas cleaning system of a waste incinerator has to cope with a number of pollutants evolving during waste incineration such as fly ash, hydrogen chloride and -fluoride, sulphur dioxide, heavy metals, and (chlorinated) hydrocarbons. These contaminants are partly reaction products from waste incineration; others are produced in the boiler itself, such as dioxins and furans (PCDD/F). The (internal) power consumption of the entire flue gas treatment process varies between 8 and 15% of the gross power production, depending on the components installed. The following section should be a brief overview about commonly applied components used in the flue gas cleaning process.

Dust and fly ash can be removed from the flue gases by electrostatic precipitators (ESP) or fabric bag filter systems. ESP uses an electric field to transfer a charge to particles, which causes the dust to stick to the oppositely charged electrode. Mechanical vibrators remove the dust layer from time to time. The removal efficiency of ESP is approx. 98% for particles sizes larger than 0.1 µm. Fabric filters consist of several separate filter chambers. The efficiency is largely improved if a dust layer has been built up blocking the passage for smaller particles. This effect also increases the pressure drop, leading to higher energy consumption. Fabric filters are mechanically cleaned by shaking or pressure pulse. The efficiency is generally higher compared to ESP (approx. 99.5% for particles larger than 0.1 micron), but they are limited in temperature (max. 160°C) and tend to have higher operational costs.

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[12] E. DIRKS: „Praxishandbuch Abfallverbrennung – Technik und Betrieb thermischer Behandlungsverfahren“, Herrentor Fachbuchverlag, ISBN 3-00-005535-5, (2000)

Scrubbing processes are commonly used to reduce the concentration of SO_2, acid components and volatile heavy metals such as mercury. In dry scrubber processes, powdered alkali such as hydrated lime (calcium hydroxide) is injected into the flue gas stream. The alkali reacts with acid gases, which produces solids that can be captured in the dust removal system. In semi-dry processes, lime is sprayed into a reactor located in front of the dust removal system. The droplets evaporate in the reaction tower. The resulting dry powder consists of calcium chloride, fluoride, and sulphate. The solid material is captured in the dust removal system. Comparable to the dry system, the removal of mercury can be improved by using carbon or chemical reagents such as sodium sulphide.

A further technology is the use of wet-scrubbing systems, capable of removing gaseous contaminants as well as dust particles. They are usually located behind the dust removal system in order to reduce fouling and wear. The wet scrubber uses a liquid absorbent such as lime to wash the flue gases. In a vertical column, several dispensers spray the liquid absorbent down onto the upstream flowing gases. In the first stage, HCl and HF are removed, as well as (residual) dust and condensed heavy metals (acid scrubber). The following stage removes sulphur oxides (neutral scrubber). The efficiency of the wet scrubber is high, leading to the lowest amount of solid residue (compared to the dry and semi-dry scrubbers), while minimal absorbents are necessary. The water from the scrubber can be drained or evaporated in order to concentrate the pollutants.

Nitrogen oxides (NO_x) are removed (or reduced to N_2) from the flue gases with DeNOx installations. Two processes are commonly applied, which are known as Selective Catalytic Reduction (SCR) and Selective Non-Catalytic Reduction (SNCR). In the SNCR process, ammonia or urea is used as a reactant and sprayed into the boiler at high temperatures (approx. 900°C) in order to convert nitrogen oxide to molecular nitrogen (N_2) and water vapour. In SCR systems, a catalyst is used to allow a reduction of NO_x at lower temperatures, while a reactant such as ammonia is still required. The location of the SCR system is important for the functioning of the catalyst. Excess dust or SO_x can clog the system and can decrease its life span. Due to this reason, SCR systems in MSWI are usually installed in low-dust setting. If the facility is equipped with a SCR system, they are often realised as so-called “combined-catalysts”, allowing the reduction of NO_x and the destruction (oxidation) of organic pollutants such as PCDD/F in two separate process steps.

A further option to reduce the pollutant concentration in the waste flue gas is the use of active cokes, which are capable to adsorb micro-organic contaminants. The powdery cokes can be added to the flue gases and have to be separated by the dust removal system (fabric filter), although this approach is not considered beneficial as the total amount of fly ash is increased. Furthermore the mixed fly ash/coke mixture is highly contaminated and has to be disposed of accordingly. An alternative method is the use of fixed bed filter. The efficiency of fixed bed filter is usually higher in comparison to coke injection with the exception of

PCDD/F. On the other hand fixed bed filters have a higher pressure drop, leading to increased operational costs. Nevertheless, the loaded coke can be extracted from the filter and fed to the combustion system, where organic pollutants are destroyed.

2.1.4 Residue treatment

A major principle of thermal waste treatment is volume reduction of waste materials. The incineration process leads to a decrease of about 90% of the original volume and to 70% of the original weight. The residues are slag from the incineration furnace, fly ash from the dust removal system, and residues from the scrubbing processes. Several residues can be reused in various applications, decreasing the total amount to be disposed of:

- Quenched slag is conveyed to a special slag bunker underneath the boiler. Iron can be separated by means of magnetic drums. The slag is inert and can be used for e.g. road construction.
- The fly ash separated in the dust removal system is temperately stored in a silo until the product can be used as an additive in asphalt mixtures or as backfilling material.
- The residues from scrubbing processes consist mainly of (contaminated) mixed-salts (after evaporation of process water). Further reuse is difficult and final deposition is - in most case - inevitable.

2.2 Solid recovered fuel (SRF) production and utilisation

From a technical point of view, state-of-the-art waste and residue treatment techniques of the last decades have been and still are recycling (reuse), incineration (thermal disposal) and the (final) disposal on landfills. Considering the costs, the most economic - and in several European Countries the only treatment path - is the disposal on landfills. As the environmental deficits (uncontrolled emissions, contamination of soils and ground water, no material or energy recovery) are no longer acceptable, the European Landfill Directive (1999/31/EC) was implemented, which is one of the major driving forces to develop and implement - aside from recycling and incineration - further environmental and economical sound alternatives in an integrated and sustainable waste treatment scenario.

The production and energetic utilisation of fuel recovered from (non-hazardous) residues and waste materials is a good example for an environmental and economical sound alternative. With increasing energy costs, but also due to necessities following the landfill directive, there is a growing interest of the industry (cement, lime, steel and energy), authorities and politicians to enhance the production of SRF produced from non-hazardous residues and waste materials. Due to liberalisation and need for cost reduction, the industry is highly interested in less expensive homogeneous substitute fuels of a specific quality.

2.2.1 Origin, production and thermal utilisation of SRF

Mixed SRF mainly consist of biogenic components (45-65 wt.-%) like paper, cardboard, textiles, and wood. A further significant fraction consists of mixed plastics - such as PE, PP, or PS - in form of foils or (hard) plastic pieces. They derive from non-hazardous mixed waste streams, such as municipal solid waste (MSW), commercial, or bulky waste, but also from (certain) mono waste streams. More specific, the input materials suitable for SRF production are defined according to the waste catalogue and the Commission Decision 2000/532/EC as the following five main groups:

- Group 1: wood, paper, cardboard and cardboard boxes
- Group 2: textiles and fibres
- Group 3: plastics and rubber
- Group 4: other materials (e.g. waste ink, spent activated carbon, etc.)
- Group 5: high calorific fractions from non-hazardous mixed wastes

The high calorific fraction (HCF) is usually sorted out from the mixed streams by positive or negative sorting technologies and mixed with defined production specific waste streams in order to achieve the required quality for the final product. The application of automatic sorting technologies such as NIR (near infrared detection and separation) enhances the quality of the product. Further process steps contain size reduction, classifying, mechanical sorting, ferrous metals (FE) or non-ferrous metals (NF) separation, biological drying, etc. The two main approaches on the SRF production contain either mechanical processing steps to separate the high-calorific fraction and to remove unwanted components such as PVC, or mechanical-biological treatment where a biological drying step is integrated in the process. SRF is usually produced in the form of bales, fluff, soft or hard pellets according to the market demand.

2.2.2 Definition of quality standards

As the number of SRF production plants grows in Europe, there is an increasing demand on efficient quality control mechanisms in the waste treatment processes. While in the previous decades, the input material for SRF was mainly product specific waste streams, the development of the sorting and separating technologies in the present time enable the utilization of mixed waste streams which are more difficult to handle and control. The need for quality assurance and fuel standardization was recognised by the SRF producers. Thus, the quality control of the input streams and the prevention of misuse or illegal disposal of hazardous waste, e.g. by dilution of critical waste streams and components in the mixed SRF, was indicated as a matter of prime importance. National regulations were developed like the Regulation of the German Institute for Quality Assurance and Certification (RAL-GZ 724). On European level the standardisation activities related to SRF are combined and coordinated in the CEN-TC 343 [13] and related national mirror committees.

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[13] J. MAIER, Th. HILBER, G. SCHEFFKNECHT: “Current activities in terms of SRF standardisation”, Conference on Waste & Biomass, Wroclaw, Poland, (2005)

2.2.3 Current and expected SRF utilisation

Actually, the main SRF users are found in the cement and lime industry. The steel industry uses SRF as a carbon-substitute and the Scandinavian countries burn SRF for district heating. Major countries producing SRF in the European Union are Germany, Italy, the Netherlands and Scandinavian countries. Currently about 1.5 Mio. Mg/a SRF are produced from MSW with a biogenic share between 45-65 % is utilised in Europe. The expected development of SRF utilisation in Germany is given in Fig. 2-3:

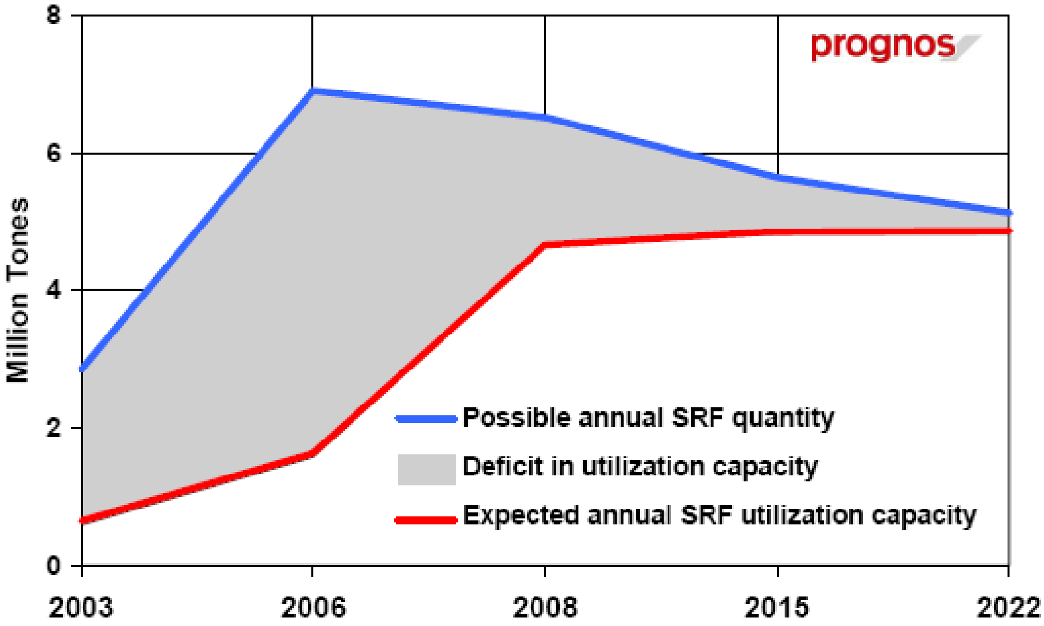

Fig. 2-3: Development of SRF quantities and capacities in Germany [PROGNOS, 2005]

The figure shows that there is a capacity deficit between possible annual production capacity and expected utilisation capacity. SRF co-combustion in existing utility boilers may play a key role at this point. Partial substitution of coal by SRF in large-scale power plants can effectively assist covering the capacities limitations to a certain extent, and will furthermore result in savings of valuable fossil fuel sources and reduction of CO_2 emissions [14]. On the economic side, SRF co-utilisation in existing thermal plants usually requires moderate additional investments.

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[14] Th. GLORIUS: “Potential for decreasing of CO_2 emissions through co-combustion of recovered fuels”, Presentation Entsorga Seminar, Cologne, Germany, (2003)

2.3 Current situation on the European waste- and power market

To assess the potential of alternative waste treatment processes such as direct SRF co-combustion or the UPSWING process, it is necessary to take a look on the actual situation. In the following section, waste potential and available qualities as well as current waste treatment routes are discussed. Of special relevance for both processes is the availability of fossil fuel-fired power plants, which has to be reviewed as well.

2.3.1 Waste potential and available qualities

Based on OECD and EUROSTAT statistics, an average waste generation of 1.4 kg/cap/d (± 0.2) has been found with a good correlation of waste generation and economical power of a country. Countries with a GDP (gross domestic product) below 10000 €/cap/y may have a generation rate as low as 0.6 kg/cap/d, whereas countries where the GDP exceeds 25000 €/cap/y produce more than two times that amount. The situation is shown in Fig. 2-4a. The net calorific value (NCV) in the EU15 countries was found to be in the order of 8.5 -11.5 MJ/kg, whereas in the eastern countries this margin stays often below 8.5 MJ/kg (see Fig. 2-4b). Furthermore, there is a surprising correlation between the GDP and the calorific value of MSW. For almost all investigated countries a biogenic fraction of the energy inventory in the waste of more than 50% could be determined.

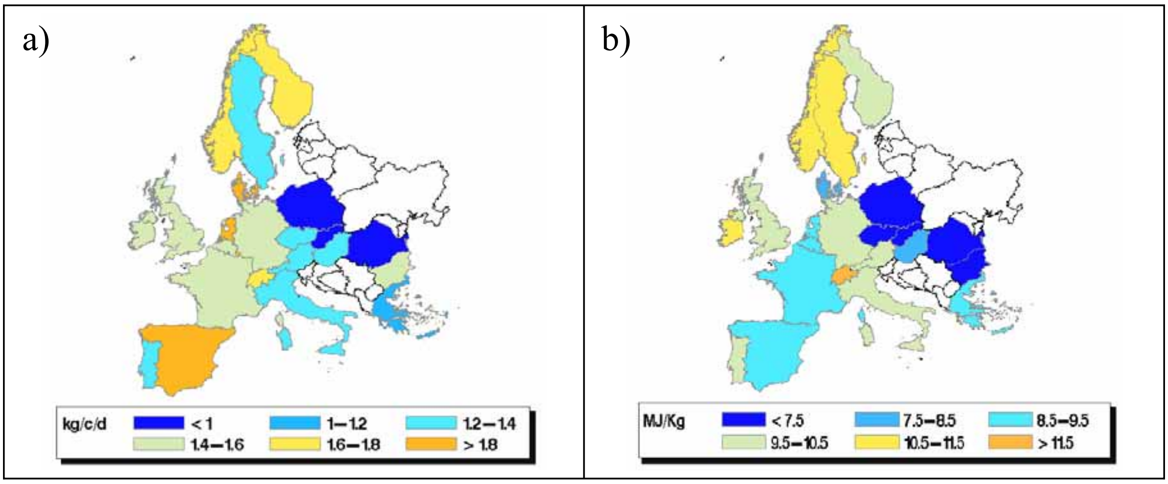

Fig. 2-4: a) Average generation of MSW, kg/cap/d b) average NCV of MSW in Europe, MJ/kg

Considering waste incineration on the grate, a sufficient net calorific value is crucial to sustain self-combustion and to guarantee a complete burnout of the waste. NCVs in the range of 9.5 to 10.5 MJ/kg are favourable, leading to low organic pollutant concentrations (CO, PAH, PCDD/F) in the waste flue gases. NCVs below 6.5 MJ/kg may require additional co-firing of fossil fuels such as oil or coal (pre-mixing). Considering the production of SRF, it has to be mentioned that low NCVs are often the result of considerable high biomass content, whereas the values often vary largely between rural and urban areas. As a consequence, the question if a production of SRF is technical and economical feasible, has to be evaluated from case to case.

2.3.2 Most important treatment and disposal routes

The waste management in Europe - regardless of the implementation of the EU Landfill Directive in 2005 and with the exception of some countries which have already issued a landfill ban for untreated waste - still mainly rely on landfill. Fig. 2-5 shows the most important treatment and disposal routes for municipal solid waste in Europe:

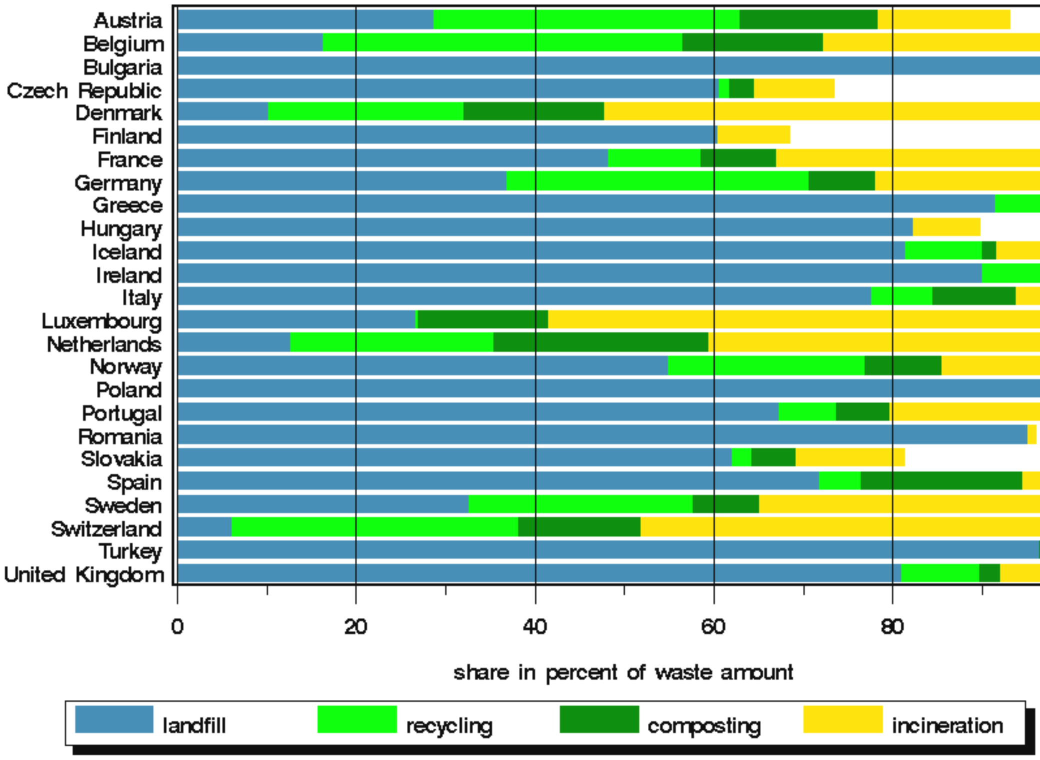

Fig. 2-5: Most important treatment and disposal routes for MSW in Europe

In the new member states of the EU, there is almost no incineration capacity installed. The same is still valid for some of the EU-15 countries like the UK, Ireland, or the Mediterranean countries, which landfill most of their waste even today. In view of the future compliance with the Waste Landfill Directive, a gradual decrease of the waste quantities disposed off to landfills [15] and a push into the direction of thermal waste treatment is expected in those states during the next years.

2.3.3 Current situation of fossil fuel utilisation in power production

In order to estimate the market chances for both UPSWING and SRF co-combustion requires an evaluation of the energy market in Europe. Both processes largely rely on the availability of fossil fuel fired power plants. It was found that the East-European countries rely heavily on fossil fuel, particularly coal. France and the Scandinavian countries (Sweden and Norway) use very little coal, having high levels of nuclear and hydro-electric power respectively. As described in the previous chapter, the UPSWING concept is especially of interest

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[15] European Commission Directorate General for Environment “Refusal Derived Fuels - Current Situation and Perspectives” (B4-3040/2000/306517/MAR/E3), Final Report, (2003)

for those countries which largely rely on fossil fuels and lack of state-of-the-art waste treatment facilities. Countries fulfilling these prerequisites are primarily found among the New Member and Associated States (Poland, Romania, Bulgaria), but also in Southern Europe (Italy, Spain, Greece).

2.4 Process-specific environmental regulations

There are different legislative regulations concerning the emission of the two processes which are combined in the UPSWING Process. The environmental standards of a power plant are regulated by the Large Power Plants Directive (Directive 2001/80/EC), whereas a MSWI has to comply with the Waste Incineration Directive (Directive 2000/76/EC). The relevant limits for both directives are summarised in Appendix D-1 including the German counterparts, the 13th and 17th Federal Imission Protection Directives (BImSchV).

At the present time, it is considered that the coal-fired power plant involved in the UPSWING Process would have to comply with the requirements of the Waste Incineration Directive, rather than those of the Large Combustion Plant Directive, because of the introduction of the incinerator flue gases to the system. Considering direct SRF co-combustion, the Waste Incineration Directive has to be applied anyway, regulating both mono-combustion of MSW in dedicated facilities and co-combustion of SRF in industrial furnaces or power plants. In power plants, the maximum share of SRF is limited to 25% of the thermal input, while the emissions limits have to be calculated according to the mixing rule.

2.5 Scope and content of this thesis

The main objective of this thesis is to qualify the UPSWING process as a potential option in a future waste management scenario. Considering the propositions of the process, to solve economical deficits of conventional MSWI while maintaining its environmental benefits, especially in comparison to alternative waste treatment concepts such as direct SRF co-combustion, it is obvious that these propositions have to be experimentally validated. However, the UPSWING process is still a theoretical concept and a direct comparison on full-scale basis is not possible yet. Hence, an experimental approach has to be found which allows a comparison of the above-mentioned processes. Chapter 3 is dedicated to this task.

For a comparison of the different processes, three important areas were identified in chapter 1.4.2, comprising of (a) environmental aspects, (b) operational aspects, and (c) economical aspects. The interrelation of UPSWING, SRF co-combustion, MSWI, and thermal power plant process (TPP) considering these aspects is schematically shown in Fig. 2-6:

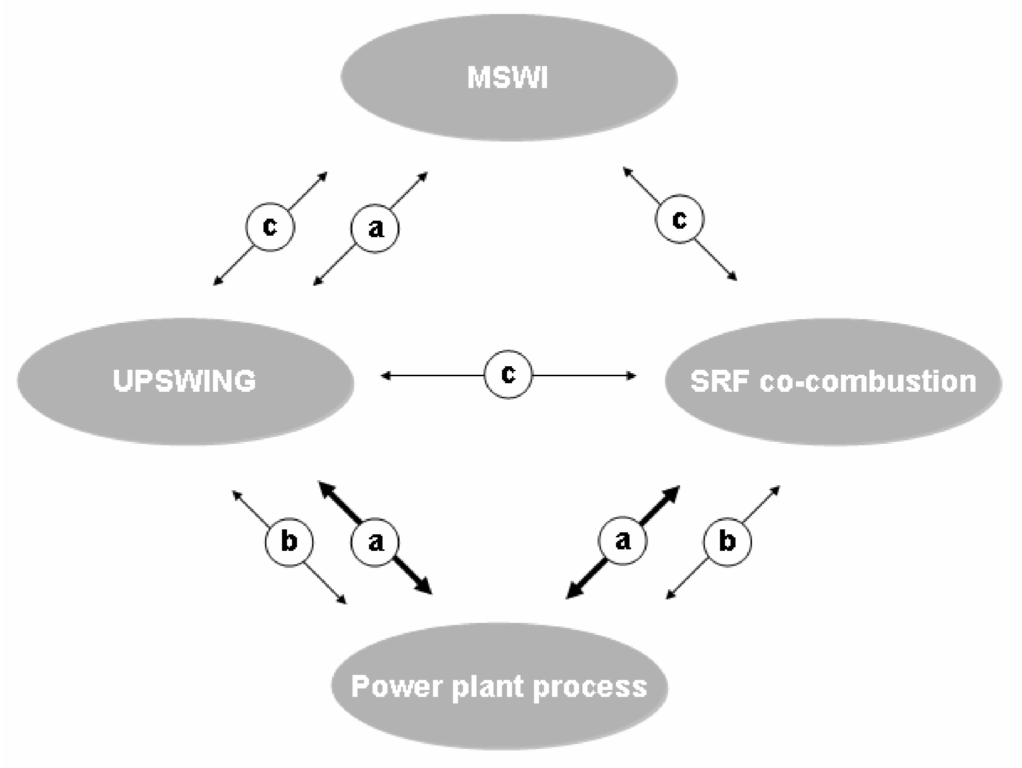

Fig. 2-6: Interrelation of the investigated processes

Considering the environmental implications, the UPSWING process proposes a comparable environmental performance to conventional MSWI. This aspect has to be validated, especially in terms of direct pollutant destruction, whereas PCDD/F should be highlighted in this context. Furthermore, the emissions of the integrated process have to be evaluated against the standalone power plant process (TPP). A comparable approach can be formulated for SRF co-combustion, while the process emissions are subject to a comparison to the standalone process, too. This entire complex is the main objective of the following chapters, and the main objective of this thesis, as well.

Considering the operational aspects of the UPSWING process, it is important to assess the impact of flue gas integration on the power plant process. Chapter 5 focuses on aspects such as corrosion potential and residue quality, and a comparable experimental approach was considered for direct SRF co-combustion. A more detailed study on flue gas integration and impact on boiler performance bases on a theoretical approach (numerical simulation), and can be found in chapter 6.

In economic terms, the UPSWING process proposes lower costs for the thermal treatment of waste in comparison to conventional MSWI. A comparable statement should be valid for direct SRF co-combustion. Economic aspects are highly relevant, of course, though not part of the experimental work, and therefore not subject of this chapter. A brief discussion of this comparison can be found in chapter 7.

3 Process verification requirements

Prior to the detailed discussion of the experimental results, which can be found in chapter 5, it is necessary to take a closer look on the processes itself and to work out the relevant priorities in the respective areas of concern (environmental/operational). This will be done for both UPSWING in chapter 3.1 and direct SRF co-combustion in chapter 3.2. Furthermore, the experimental boundary conditions will be briefly presented.

3.1 The UPSWING process

The UPSWING concept describes the combination of a waste incineration facility with a conventional power plant. Combination is generally realised by either steam- or flue gas integration or - in case of the UPSWING process - both options. The major principle behind the concept is to keep problematic pollutants consequently away from the power plant. To achieve this, cleaning or partial cleaning of the waste flue gases from the grate firing system is necessary prior to injection into the power plant process.

The impact of waste flue gas integration on the power plant process (operational) and the emission behaviour (environmental) has been investigated in detail. The results are summarised in chapter 5 and 6. Hence, it is necessary to characterise the properties of the waste flue gases introduced into the power plant process. Based on these findings, priorities (What is important in this context? What has to be investigated?) could be defined, accordingly. Consequently, the experimental setup should be capable to answer these questions.

3.1.1 General characterisation of waste flue gases

In chapter 2.1, the conventional waste incineration process including its main components was briefly introduced. As mentioned, the grate firing concept including the boiler system required for the UPSWING process is similar to those used in conventional MSWI. Consequently, comparable raw flue gas concentrations can be expected for the UPSWING grate firing system.

The combustion process on the grate is characterised by high temperatures and a significant oxygen surplus. This leads to an almost complete conversion of all input species into stable products. Furthermore, a partitioning between species staying in the fuel bed and species entering the flue gas path takes place. The primary combustion products are given in Fig. 3-1. Mineral materials as well as lithophilic heavy metals like iron (Fe), manganese (Mn), titanium (Ti), nickel (Ni), or copper (Cu) and their compounds leave the grate mainly with the bottom ashes. A certain amount of salts like alkali or earth-alkali chlorides and sulphates stay in these residues as well.

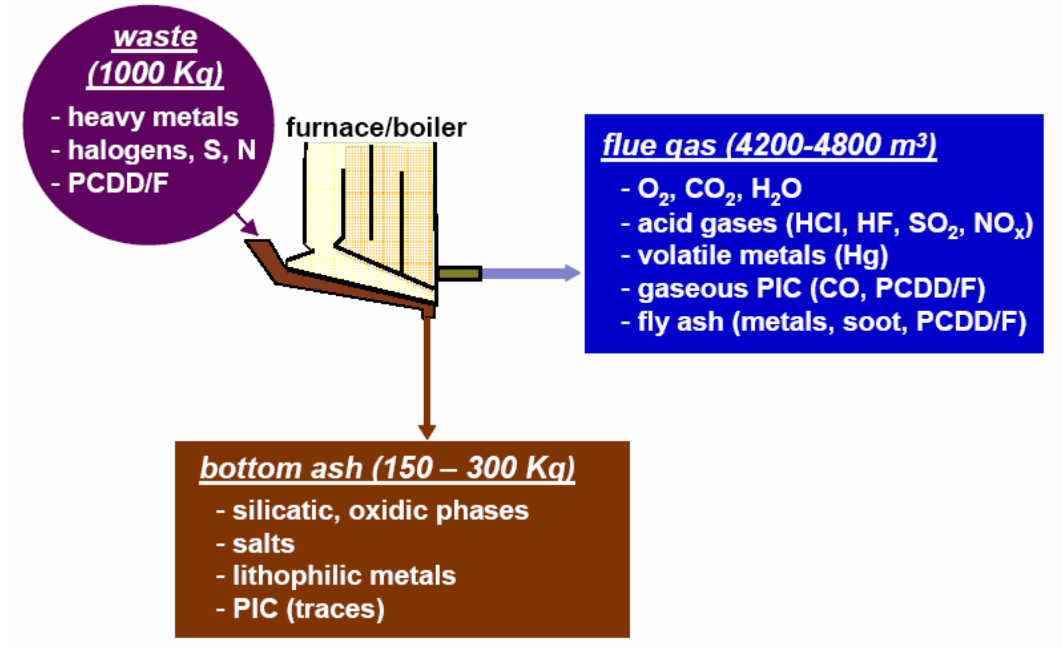

Fig. 3-1: Primary combustion products and partitioning in a waste incineration facility [16]

Sulphur may form the gaseous SO_2, though the formation of sulphates and the removal with the bottom ash is likely. The waste bound nitrogen is partly oxidised to NO with small amounts of other oxides like NO_2 and N_2O. The oxidation of air nitrogen (thermal NO) has no relevance since the combustion temperatures are too low. Halogens (Cl, F, Br) are transformed into the respective halides. In case of Cl the major product is HCl, whereas only 10-15% stay as chlorides in the solid residues. Volatile heavy metals are mainly evaporated as chlorides. Almost all mercury (Hg) and approx. 80% of the inventory of thallium (Tl) and cadmium (Cd) leave the combustion chamber with the raw flue gases. Other elements like zinc (Zn), arsenic (As), antimony (Sb), and lead (Pb) are almost equally distributed between bottom ash and raw gas. At the boiler exit (180-220°C), however, all volatile heavy metal species but Hg and its compounds are bound to particulate matter and are discharged with the filter (fly) ashes [17].

Since the oxidation of the carbon inventory results not completely in CO_2, minor amounts of incomplete combustion products such as carbon monoxide (CO), poly-aromatic hydrocarbons (PAH), or particulate carbon are still present in the gas phase. The latter one is known to be a source of toxic low-volatile organic micro-pollutants, especially of PCDD and PCDF. According to Vogg [17] PCDD/F are predominantly formed by a heterogeneous oxy-chlorination reaction, the de-novo synthesis, inside the boiler. The improvement of the waste

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[16] J. VEHLOW, H. HUNSINGER, S. KREISZ, H. SEIFERT, Th. HILBER, J. MAIER, K.R.G. HEIN: "Increasing the Eco-efficiency of Waste Incineration with the UPSWING Process", IT3'05 Conference, May 9-13, 2005, Galveston, TX. Proceeding on CD, document IT3-43, (2005)

[17] H. VOGG, L. STIEGLITZ: "Thermal Behaviour of PCDD/PCDF in Fly Ash from Municipal Incinerators", Chemosphere, 15, 1373, (1986)

burnout during the last years resulted in a drastic reduction of PCDD/F raw gas concentration. Today's state-of-the-art waste incineration facilities have raw gas concentrations in the range of 1 ng (I-TEQ)/m³ [18]. The following table summarises typical pollutant concentrations in the raw gases of a typical grate firing system. The given ranges represent the usual fluctuations in the composition of the waste [19]:

Table 3-1: MSWI raw gas pollutant concentrations

Pollutant	Raw gas concentration		Unit
-	Min	Max	11% O_2 (dry)
O_2	5	8	%
N_2	66	72	%
CO_2	8	13	%
Dust	1000	5000	mg/m³
HCl	500	2000	mg/m³
SO_2	150	400	mg/m³
H_2O	110	150	g/m³
NO_x	100	500	mg/m³
NH_3	5	30	mg/m³
CO	< 10	30	mg/m³
TOC	1	10	mg/m³
Hg	100	500	µg/m³
PCDD/F	0.5	5	ng(I-TEQ)/m³

3.1.2 Partial flue gas cleaning and residual pollutant concentration

In order to reduce the risk of boiler corrosion and not deteriorating the quality of the power plant's solid residues the raw gases from the waste incinerator have to be cleaned prior to injection. The UPSWING process foresees de-dusting, preferably by a fabric filter, and a wet acid scrubber stage (pH = 1). This procedure guarantees the almost total removal of particle bound heavy metals and PCDD/F, more than 98% of HCl/HF, and approx 90-95% of the total mercury input with a high efficiency for $HgCl_2$. Gaseous metallic mercury (Hg^0), which is not water-soluble, but also gaseous PCDD/F, SO_2 and NO_x, are not or only to a minor extent removed in the scrubber. Typically, the scrubbers are equipped with one- or multi-stage packing to increase the specific surface and enhance the mixture between flue gas and washing fluid [20].

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[18] G. REECK, W. SCHRÖDER, G. SCHETTER: "Zukunftsorientierte Abfallverbrennung in der MVA Ludwigshafen", Müll und Abfall, 23, 661, (1991)

[19] J. VEHLOW, H. HUNSINGER: „Identification of Flue Gas Composition Range of a Waste Incinerator", UPSWING Project, http://www.eu-projects.de/upswing, (2003)

[20] E. DIRKS: „Praxishandbuch Abfallverbrennung – Technik und Betrieb thermischer Behandlungsverfahren", Herrentor Fachbuchverlag, ISBN 3-00-005535-5, (2000)

As mentioned the partially cleaned gas still contains SO_2, NO_x, PCDD/F, and, with a significant lower concentration in comparison to the raw flue gases, HCl and Hg. Fly ash (or dust) is almost completely removed behind the scrubber. The following table summarises the expected pollutant concentration behind the partial flue gas cleaning process [20]:

Table 3-2: Pollutant concentrations of MSWI flue gas behind partial flue gas cleaning

Pollutant	Behind acid scrubber		Unit
-	Min	Max	11% O_2 (dry)
NO_x	100	500	mg/m³
SO_2	150	400	mg/m³
HCl	10	40	mg/m³
PCDD/F	0.5	5	ng(I-TEQ)/m³
Hg	5	25	µg/m³

Considering the residual pollutants in the pre-cleaned waste flue gases prior to injection, the necessity to investigate their fate (and behaviour) in the power plant process is obvious. Consequently, the experiments performed regarding the UPSWING process focus on these pollutants, viz.

Nitrogen oxides (NO_x): Nitrogen oxides are produced by coal combustion as well. Usually, the concentrations produced by coal are significantly higher. In modern coal boilers, different measures are undertaken to reduce these pollutants to N_2 directly in the combustor (in the following referred to as “Low-NOx Technologies”). Hence, a reduction of the additional NO_x freight brought in by the waste flue gases should be possible if the waste flue gases are injected at a suitable location. It is therefore necessary to investigate this basic assumption. Furthermore, the identification of the best-suited injection location for waste flue gas injection is targeted as well.

Sulphur dioxide (SO_2): The SO_2 concentration of the waste flue gases is, in comparison to those of flue gases from coal combustion (partly >> 1000 mg/m³), considerable low. SO_2 produced by the coal is commonly removed in flue gas de-sulphurisation systems (FGD). It is therefore expected that is should be generally possible to remove the additional SO_2 input down to levels which meet the required emission limits.

Hydrogen chloride (HCl): Comparable to SO_2, residual HCl concentrations behind the acid scrubbing stage are lower in comparison to those of coal (chlorine emissions from coal combustion are usually found in the range of 50-120 mg/m³). Dilution of the flue gases from coal combustion can be expected as well. Nevertheless, chlorine may influence the behaviour of other elements such as heavy metals or the properties of power plant residues such as fly ash.

Dioxins/Furans (PCDD/F): PCDD/F emissions from coal-fired power plants are extremely low [21]. Consequently, the fate of PCDD/F additionally brought into the power plant process by the waste flue gases is of special relevance, although it is expected that PCDD/F are destroyed in the combustor. Hence, the verification of this assumption is one of the most important aspects.

Mercury (Hg): As discussed before, a small share of mercury may pass the acid scrubber in form of metallic Hg^0. Coals contain mercury as well, and the usual concentrations found in coal flue gases are more or less comparable to those found in the pre-cleaned flue gases [22]. Hence, increased emissions cannot be expected. Nevertheless, the fate of mercury was investigated as well due to its environmental relevance.

3.1.3 Experimental setup and investigated fuels

The impact of flue gas integration on the power plant process was investigated in detail. The experiments were primarily conducted at a small combustion reactor, in the following referred to as BTS. The BTS is an electrically heated drop-down furnace, which is designed for the combustion of pulverised coal and subsequent scientific investigations. The reactor is equipped with various access ports and hatches, allowing the injection of waste flue gases at different locations, and therefore offering a good controllable environment for the planned investigations. A detailed description of this test facility can be found in chapter 4.1. In addition, several experiments concerning the destruction of PCDD/F have been performed at the fluidised bed reactor ELWIRA. A detailed description of this facility is given in chapter 4.3.

A solution had to be found how to inject a controllable amount of different waste flue gas qualities into the test facilities. The use of a gas mixing station was considered as the favourable option to produce artificial flue gases with reliable parameters. In addition to the simple gas mixing process, the device had to be capable to evaporate water (and other solvents such as HCl, NO_2, or SO_2) to achieve a comparable composition as outlined in Table 3-1 and Table 3-2. A detailed description of this gas mixing station is given in chapter 4.4.

With the availability of the gas mixing station, artificial waste flue gases could be generated and directly injected into the BTS combustion reactor. Two principal injection variants were investigated during the trials: Flue gas injection into the secondary air inlet of the burner (option 1) and the injection via a lateral access port in 0.9m distance from the burner (option 2). An illustration of both options is given in Fig. 3-2a:

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[21] H. HAGENMAIER, R. BEISING: "Untersuchung von Kraftwerksrauchgasen auf polychlorierte Dibenzodioxine und Dibenzofurane", VGB Kraftwerkstechnik, 69, 1024, (1989)

[22] J. VEHLOW, H. HUNSINGER, S. KREISZ, H. SEIFERT: „Das UPSWING Verfahren - Der Schlüssel zur kostengünstigen Abfallverbrennung", ITC Forschungszentrum Karlsruhe, (2000)

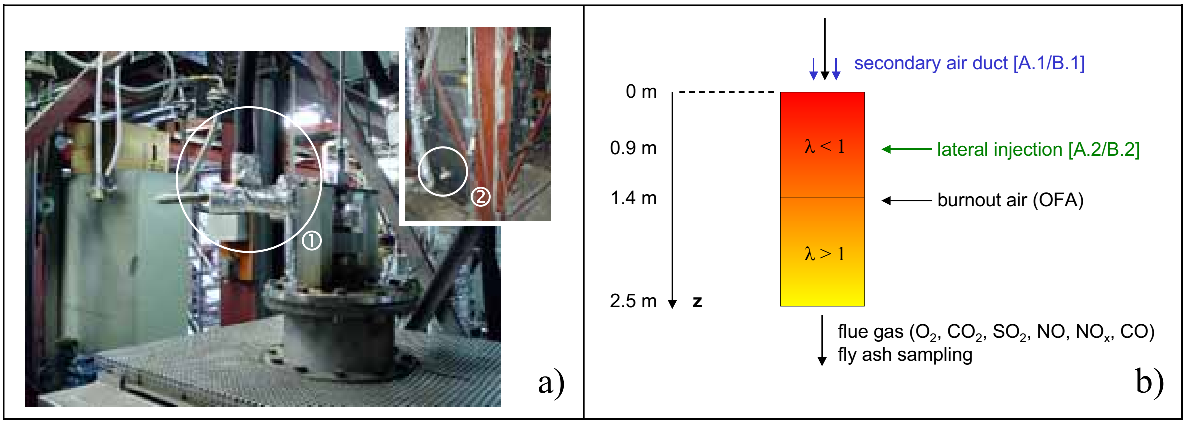

Fig. 3-2: a) Flue gas injection at test facility b) Air staging configuration

The first option corresponds to the injection of the incinerator flue gases via the burner air system respectively the vapour nozzles of a lignite-fired power plant. The second location corresponds to the injection via dedicated injection nozzles above the burner level prior to the injection of the OFA. All experiments were performed with an excess air ratio of 1.20, corresponding to 3.5% O_2 (dry) in the flue gas at the reactor exit. Emission values given in the context of this thesis are always correlated to 6% O_2 under dry conditions.

A further important aspect of waste flue gas integration into the power plant process is the oxygen content of the waste flue gases. Once the waste flue gases are injected into the combustion chamber, the oxygen level will be increased if the power plant control system is not equipped with an O_2 controller. In the following discussion this additional injection without (automatic) reduction of combustion air will be investigated and referred to as case A. If the power plant control system is equipped with an O_2 controller, the combustion air will be decreased and the remaining oxygen in the waste flue gases will be used as oxidising medium for coal combustion. Following this approach, the available oxygen in the furnace - and consequently the stoichiometry - is constant. To achieve this, the combustion air (or secondary air) was reduced during the trials depending on the injected waste flue gas amount and oxygen content. This basic operational setting will be investigated and referred to as case B.

The consequences of both operational settings were primarily investigated within the complex NO_x reduction and NO_x reduction potential, which is discussed in chapter 5.1. Furthermore, the influence of flue gas integration on primary NO_x reduction measures was investigated for different air ratios below stoichiometric conditions in the main combustion zone. Burnout air was injected at two different positions from the burner (L = 0.9 and 1.4m) in order to realise different residence times under reducing atmosphere (see Fig. 3-2b). Excess air ratio behind burnout air injection was re-adjusted to 1.20 (3.5% O_2 dry) in order to achieve full fuel burnout.

The impact of waste flue gas integration on the coal combustion process was investigated for three different coals, a Colombian bituminous coal (B/1), a Polish lignite (L/1), and a Czech hard brown coal (L/2). The bituminous coal is a typical import coal, combusted in many multi-utility boilers throughout Europe, and therefore selected as a representative base fuel for the investigations. Both lignite and hard brown coal are coals combusted at the respective power plant sites of two project partners taking part in the European Research Project UPSWING. Detailed analytical data including the results from proximate, ultimate, and elemental analysis is summarised in Appendix A-1. The specific flue gas volumes, the specific air demand, the net calorific value (NCV), and the ash content of the coals are summarised in the following table. The fuel properties do not show the raw analytical data as all fuels had to be pre-dried and pulverised prior to their application in the test facilities. Accordingly, the given data characterise the coals “as combusted”. This approach is valid for the entire thesis.

Table 3-3: Coal characterisation UPSWING

Parameter / Coal	Col. bituminous coal , B/1	Polish lignite L/1	Czech hard-brown coal , L/2
Water content, %	2.4	4.2	4.2
Ash content, % dry	15.5	35.5	19.7
NCV, MJ/kg	25.965	15.689	19.468
Nitrogen (N), % daf	1.5	0.6	0.9
Sulphur (S), % daf	0.6	1.0	0.9
Chlorine (Cl), % daf	0.05	0.06	0.06
Specific air demand, m^3/kg	6.84	4.33	5.84
Specific dry flue gas volume, m^3/kg	6.59	4.16	5.66

In order to achieve comparable conditions during the trials, the waste flue gas volume injected into the test facilities was calculated based on the flue gas volume generated by coal combustion. This ratio, expressed as the amount of waste flue gases divided by the amount of flue gases from coal combustion, was adjusted between 10 and 20%. As an example, the additional injection of waste flue gases on a ratio of 10% results in 0.659 m^3/kg_{coal} for the bituminous coal. This ratio has to be multiplied by the excess air level (in case of the BTS trials multiplied by 1.2). Furthermore, this waste flue gas ratio can be theoretically related to the initial heat input of MSW on the grate firing system (thermal share $MW_{waste}/MW_{(waste+coal)}$). To allow this, flue gas balances were calculated for an average MSW with a net calorific value of 8680 kJ/kg, generating approx. 4.6 m^3/kg waste flue gas at an air ratio of 1.6. The results of these calculations are summarised in appendix B-1, and resulted in an average thermal share of 5-6% for a 10% flue gas ratio and 10-12% for a 20% flue gas ratio (see Appendix B-1, figures B-1/1 through B-1/3).

3.2 Direct SRF co-combustion

SRF co-combustion describes the direct co-combustion of pre-processed waste materials in coal-fired boilers or industrial furnaces. Pre-processing covers the production and the pre-treatment of a fuel to such an extent allowing its direct utilisation in a power plant boiler. Waste specific pollutants such as chlorine or heavy metals need to be minimised during the fuel production process. The utilisation of high calorific and relatively clean fractions such as paper, cardboard, or plastic should be favoured. Nevertheless, it should be clear from an economic, but also from a technical point of view, that the reduction of pollutants has its limits. Taking this into account, it is clear that the residual pollutant concentrations in SRF materials may exceed the concentrations usually found in coal by several magnitudes. As a consequence, direct SRF co-combustion increases the concentration of specific pollutants in the power plant process. Heavy metals are an example, but also chlorine is problematic, and this may result in operational as well as environmental problems. Hence, the investigation and evaluation of the direct co-combustion concept towards its impact on the power plant process is considered highly relevant, both from an environmental and operational point of view.

In order to evaluate this potential it is necessary to characterise the properties of the investigated SRF materials. Based on these results, it is possible to work out the differences in comparison to the UPSWING process, and to define priorities for the experimental programme, accordingly. These priorities may deviate slightly from the priorities defined for the UPSWING process due to the different nature of both concepts.

3.2.1 Characterisation of SRF materials

The SRF co-combustion concept has been subject to several experimental trials ranging from lab- to large-scale application. The following materials have been used during these experiments, and their differences should be elaborated in the following section:

- **SRF/04**: SRF quality produced from high calorific fractions (HCF) out of MSW (75 wt.-%), mixed with 25 wt.-% commercial waste to reduce the chlorine content
- **SRF/05**: SRF quality produced from HCF out of MSW
- **SRF/PRR**: SRF quality produced from paper recycling residues including mixed plastics, paper, and textiles.

Detailed analytical data including the results from proximate, ultimate, and elemental analysis is summarised in Appendix A-2. To give a brief impression of the fuel properties, the specific flue gas volumes, the specific air demand, the net calorific value (NCV), and the ash content of the SRF materials are summarised in the following table. Comparable to the analytical data shown in the previous chapter, the given data shows the fuels “as combusted”.

Table 3-4: SRF characterisation

Parameter / SRF	SRF/04	SRF/05	SRF/PRR
Water content, %	2.6	2.5	3.2
Ash content, % dry	12.9	11.5	8.3
NCV, MJ/kg	22.851	20.768	22.796
Nitrogen (N), % daf	0.78	0.95	0.20
Sulphur (S), % daf	0.10	0.20	0.05
Chlorine (Cl), % daf	0.34	0.64	1.13
Aluminium (Al_2O_3), % ash	45.9	17.3	19.8
Mercury (Hg), mg/kg ds	0.10	0.16	0.20
Specific air demand, m³/kg	5.73	5.09	6.29
Specific dry flue gas volume, m³/kg	5.42	4.87	5.96

In comparison to usual concentrations found in coals (see Table 3-3), the sulphur content of all materials is considerably low. Consequently, lower SO_2 emissions can be expected during SRF co-combustion. Nevertheless, there are significant differences between the materials, viz.

- SRF/PRR shows a low nitrogen content (0.2 wt.-%$_{daf}$); both SRF/04 and SRF/05 have a significant higher amount of fuel nitrogen ranging between 0.8 and 1.0 wt.-%$_{daf.}$ In case of SRF/PRR co-combustion, one can expect significant lower NO_x emissions. An assessment of this potential benefit is therefore necessary.
- The chlorine content of SRF/PRR is considerably high (1.13 wt.-%$_{daf}$), whereas the chlorine content of SRF/04 is very low (0.34 wt.-%$_{daf}$) for a waste-derived fuel. The chlorine content of SRF/05 is found in-between and was determined to 0.64 wt.-%$_{daf}$. In comparison to coal, the chlorine concentrations of all SRF materials are significantly higher. Therefore, increased chlorine emissions can be expected during co-combustion, which has to be investigated, accordingly.
- The aluminium content of the SRF/04 is considerably high, which can be correlated to a significant amount of metallic aluminium in the raw material. This can be explained with the use of commercial waste and DSD material in the SRF production process. Hence, increased aluminium content in the mixed co-combustion ashes can be expected.
- The mercury content of the investigated SRF materials ranges between 0.10 and 0.20 mg/kg in the dry substance (ds).

Regarding the ash composition (major and minor ash elements) in comparison to those of coal, SRF usually contain lower amounts of silicon (Si) and aluminium (Al) and higher amounts of calcium (Ca). Furthermore, the total alkali content is slightly higher, which may result in a stronger tendency towards slagging and fouling. Considering the trace element content in the SRF materials, significant differences can be observed to those of coal. Higher concentration can be determined for copper (Cu), lead (Pb), cadmium (Cd), chromium (Cr), nickel (Ni), and zinc (Zn). However, the element concentrations of mercury (Hg) and arsenic (As) are comparable or even lower to those of coal.

In order to achieve comparable conditions during the trials, the mass flow of SRF injected into the test facilities based on a specific thermal share. This means that a certain amount of coal (as the base fuel) is replaced by an additional fuel (SRF). The required mass flows were calculated for a fixed thermal input into the combustion facilities, based on the NCV of both coal and SRF. As an example, the thermal input into the process is 300kW and a thermal share of 10% SRF is envisaged. Hence, 30kW have to be provided by SRF and 270kW by the coal. In case of SRF/04, this would result in approx. 4.73 kg/h, considering a NCV of 22850kJ/kg. The required mass flow of the coal has to be calculated following the same procedure.

3.2.2 Experimental setup and investigated fuels

The impact of direct SRF co-combustion was investigated at two different test facilities, namely the already introduced BTS and the semi-technical combustion facility KSVA, which was designed to imitate the conditions in pulverised fuel fired boilers as far as possible (refer to chapter 4.2 for details). Contrary to the electrically heated BTS, the KSVA allows higher mass flows of both coal and SRF, and a thermal input up to 500kW is possible without the support of electrical heating systems. Starting from baseline trials (100% coal combustion) in order to provide reliable data for a process comparison, coal was successively replaced by SRF up to a thermal share of 10%. The experiments performed are summarised in the following table:

Table 3-5: Co-combustion experiments and applied thermal SRF shares

Base fuels	SRF/04 *	SRF/05 *	SRF/PRR **
Bituminous coal B/1	5 and 10%	-	-
Brown coal L/3	5 and 10%	5 and 10%	-
Bituminous coal B/2	-	-	5 and 10%
Bituminous coal B/3	-	-	5 and 10%
* experiments were performed at BTS (see chapter 4.1 for details) ** experiments were performed at KSVA (see chapter 4.2 for details)			

Aside from the Columbian bituminous coal (B/1) utilised during the experiments within the scope of the UPSWING process, two German bituminous coals (B/2 and B/3) and one German brown coal (L/3) were used during the co-combustion trials. Detailed analytical data including the results from proximate, ultimate, and elemental analysis is summarised in Appendix A-1. The specific flue gas volumes, the specific combustion air, the net calorific value and the ash content of the investigated coals is summarised in the following table. Again, the given data shows the fuels “as combusted”.

Table 3-6: Coal characterisation co-combustion

Parameter / Coal	German brown coal , L/3	German bituminous coal , B/2	German bituminous coal , B/3
Water content, %	4.9	1.2	1.0
Ash content, % dry	10.9	8.3	5.4
NCV, MJ/kg	21.146	30.836	32.438
Nitrogen (N), % daf	0.6	1.7	1.5
Sulphur (S), % daf	0.4	0.7	0.7
Chlorine (Cl), % daf	0.05	0.36	0.19
Mercury (Hg), mg/kg ds	0.14	0.24	0.25
Specific air demand, m^3/kg	5.51	8.17	8.53
Specific dry flue gas volume, m^3/kg	5.36	7.85	8.19

Compared to the lignite, but also to the Colombian bituminous coal B/1, both German bituminous coals B/2 and B/3 show a significant chlorine content. This aspect will be of further relevance assessing the results for chlorine found in the gas phase. Considering the mercury content, the concentrations found in the investigated coals are partly higher compared to the SRF materials. In order to evaluate the results on a common basis and as a prerequisite for the discussion in chapter 5, basic input/output balances have been calculated for the different fuels following the equations presented in Appendix B-0. The results of these calculations are summarised in Appendix B-2.

3.3 Summary and resulting chapter organisation

In this chapter, the necessary background for the process verification has been discussed in detail for both UPSWING and direct SRF co-combustion. In accordance with the initial idea of this thesis - the comparison of both processes - priorities could be defined for both processes, accordingly. In terms of the UPSWING process, the impact of flue gas integration on the power plant process, in particular the NO_x reduction potential, which is discussed in chapter 5.1, and PCDD/F destruction potential, discussed in chapter 5.3, were identified as the most important factors for a successful application. Furthermore, the impact of flue gas integration on trace element behaviour (chapter 5.4) will be investigated. The investigations towards SRF co-combustion have to focus on chlorine, which is discussed in chapter 5.2, trace elements (chapter 5.4), and the influence of co-combustion on power plant residues, particular on fly ash quality and its further utilisation as a consequence of the additional ash brought into the process by the SRF materials (chapter 5.5).

4 Experimental equipment

Subject of the previous chapter was a detailed review of the background required for the process verification. Partly, references to the test facilities used were already given. The following chapter will give a more detailed insight of the different facilities. Furthermore, modifications which become necessary during or in preparation of the trials will be highlighted. Chapters 4.1 through 4.3 describe the test facilities, while chapter 4.4 is dedicated to the gas mixing station utilised to generate an artificial waste flue gases as mentioned in the previous chapter. Chapter 4.5 and 4.6 gives a brief overview of the applied mercury and chlorine measurement technology.

4.1 Electrically heated combustion reactor (BTS)

The electrically heated combustion reactor is the core facility used during all experimental investigations. The following figure shows a schematic overview:

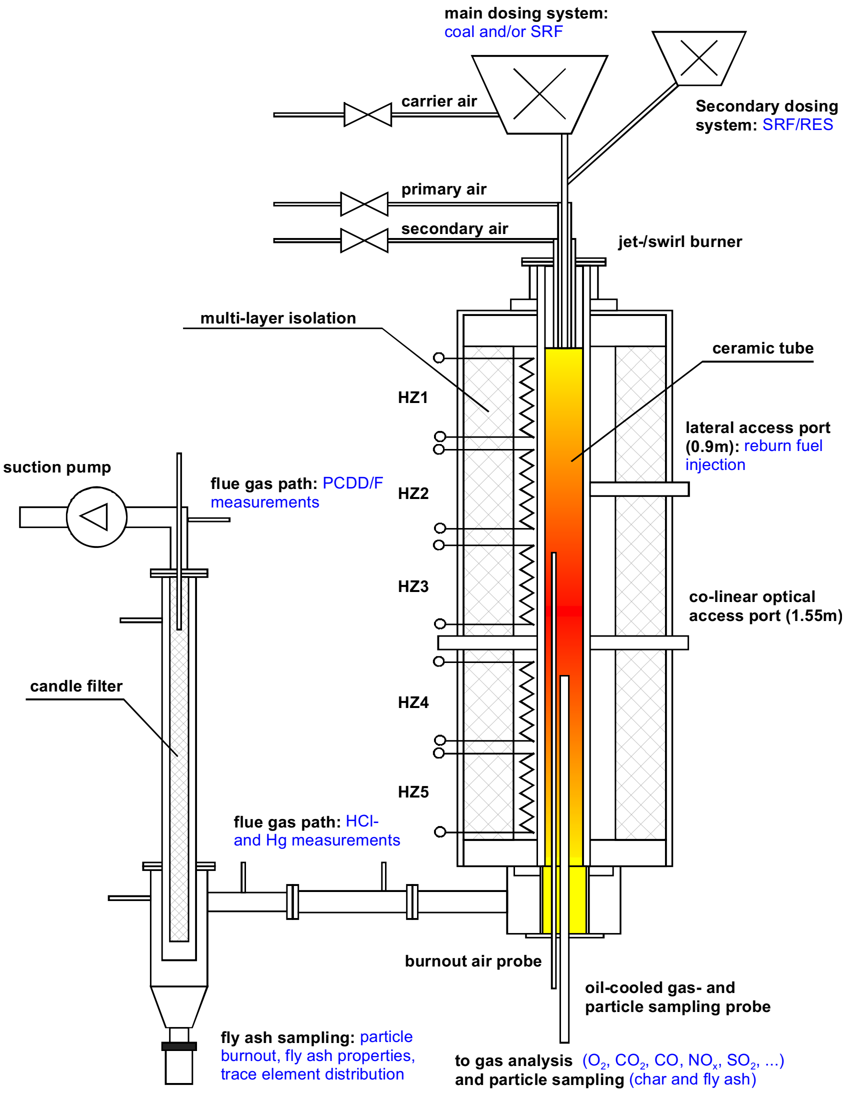

Fig. 4-1: Electrically heated combustion reactor (BTS)

The main part of the reactor is an electrically heated ceramic tube with a usable length of 2500 mm and a diameter of 200 mm. The electrical heating with an overall power of maximum 57kW allows the adjustment of a constant wall temperature as well as a temperature profile along the furnace. This enables reliable investigations of a variety of temperature-related combustion parameters from 900°C up to 1400°C. Further technical data such as dimensions, burner design, etc. can be found in [23].

4.1.1 Dosing and fuel mixing

Pulverized coal is supplied by carrier air to the top-mounted burner through which it is injected into the combustion chamber. The feeding system consists of a volumetric conveyor and a screw feeder, allowing controlled mass flows of 1 to 5 kg/h. The different air streams are continuously measured by mass flow controllers (MFC). All data are recorded and processed by a data acquisition system located next to the facility, including the information obtained from the online gas analysers. For the co-combustion trials, a possibility had to be found to inject a secondary fuel into the coal duct. A solution was found in a secondary dosing system, which allows the feeding of pre-crushed SRF material and its injection into the coal duct in front of the burner.

4.1.2 Gas sampling and analysis

Flue gas is extracted at the final section of the heated reaction tube. Standard emissions analysed are O_2, CO_2, CO, SO_2, NO and NO_x. Profile measurements of the flue gas composition can be taken by means of an oil-cooled sampling probe which transports the extracted flue gas to the standard analysers. A description of the different analysers is included in Appendix D-2. The oil-cooled sampling probe can be adjusted along the central axis of the reactor from the burner mouth to the burnout section with high flexibility. At 0.9m and 1.55m distance from the burner, lateral ports allow accessing the reaction area, e.g. for the injection of waste flue gases during the UPSWING trials.

4.1.3 Char and fly ash sampling

To characterise the volatilisation, char and emission formation, char and fly ash particle samples can be collected from the near burner field to the burnout section by the vertical adjustable isokinetic oil-cooled suction probe for further analyses parallel to the measurements of the flue gas composition. The residues collected in a cyclone sampling device during the combustion experiments can be used as fly ash samples to evaluate the impact of different combustion parameters on fly ash quality and thus their effect on flue gas cleaning devices.

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[23] U. GREUL: “Experimentelle Untersuchungen feuerungstechnischer NO_x – Minderungsverfahren bei der Kohlestaubverbrennung”, Page 45ff, VDI Forschrittsberichte Nr. 388, ISBN 3-18-338806-55

4.1.4 Fly ash removal

In preparation of the trials it became obvious that the existing fly ash removal system had to be replaced as the existing bag filter system did not allow sampling under controlled conditions. To overcome this deficit, a heated candle filter was installed, equipped with multiple sampling ports as indicated in the following figure:

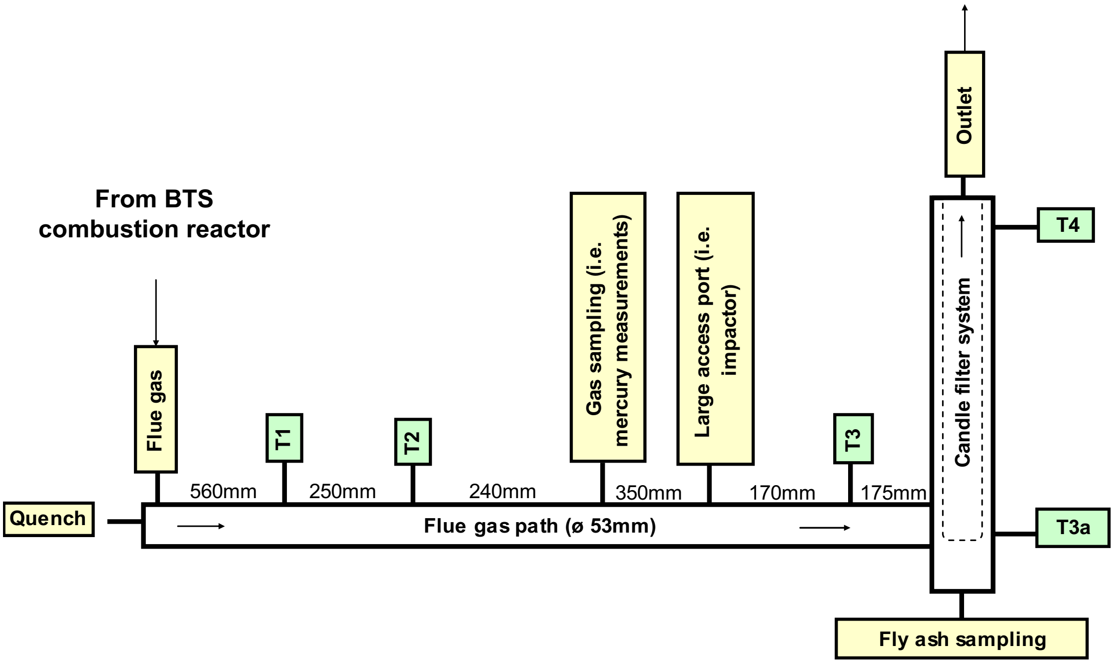

Fig. 4-2: Access ports, sampling positions and temperature measurements in the flue gas path

Furthermore, the complete flue gas path had to be reengineered to allow mercury measurements in front of the newly installed candle filter. A gas/water injection (quench) was installed to adjust the flue gas temperature in order to achieve comparable test conditions.

4.2 Semi-technical combustion facility (KSVA)

The semi-technical combustion facility was designed to imitate the conditions in pulverised fuel fired boilers. The vertical combustion chamber, top-fired and 7 m in length, is the core of the facility. The first section of 4 m is refractory lined and water-cooled (radiative part, diameter 0.75 m); the following 3 m are only refractory lined (convective part, diameter 0.75 m). The maximum residence time in the refractory lined part is 3.5 seconds. The chamber is well accessible by measurement ports which are staggered each at 90° intervals. These ports could be used for in-flame measurements (particle, temperature, and gas concentration) and also for in-furnace air and fuel staging. The flue gas path of the combustion facility consists of an air pre-heater (APH), a SCR catalyst, an electrostatic precipitator (ESP), and a fabric filter system (FF). A flow chart of the facility is given in the following figure:

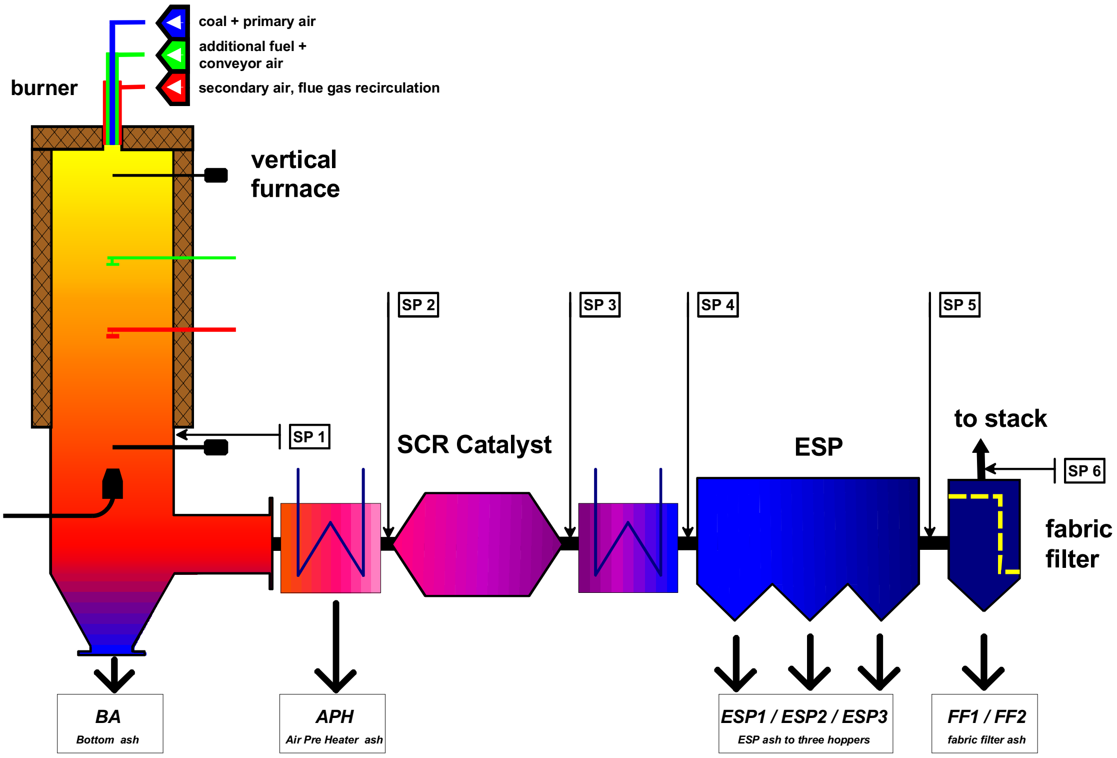

Fig. 4-3: Flow diagram of 500kW combustion facility (KSVA)

4.2.1 RES/SRF dosing and injection

The burner utilised in the tests on the KSVA is shown in Fig. 4-4a. The secondary air [1] is injected in stages and slightly swirled by means of a movable block. Coal is added via the primary air flow by the adjacent annular clearance [2]. The central element of it is the gas gun with nine gas nozzles [3] which is used for heating up and for applying a backup flame. If the coal flame is stable, the gas gun can be removed and replaced by a nozzle for SRF/RES injection. Secondary fuels can be dosed using a dedicated dosing system (see Fig. 4-4b).

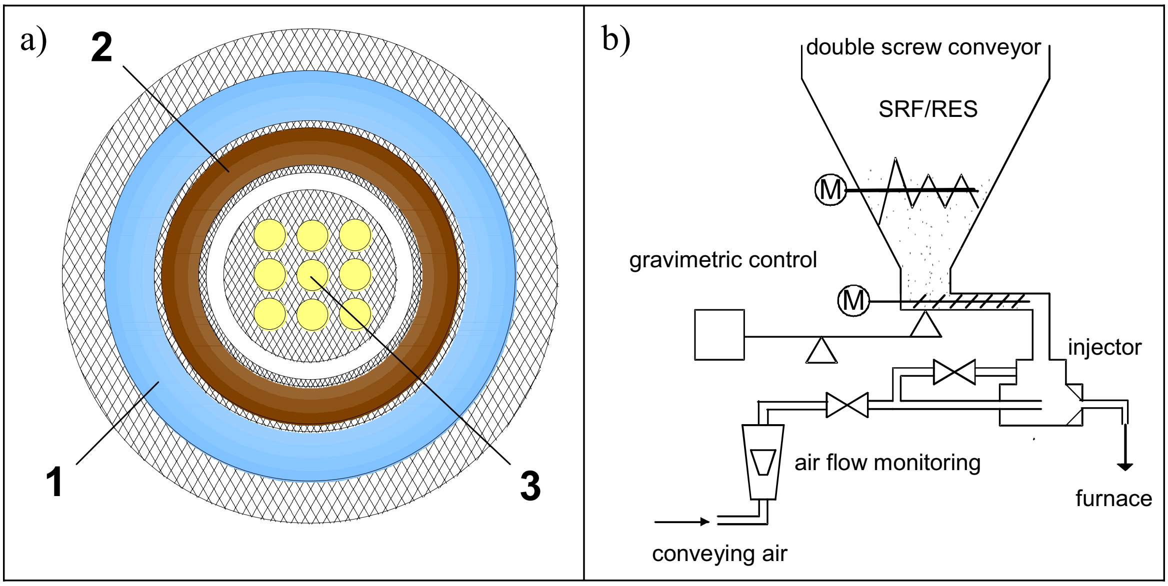

Fig. 4-4: a) Burner mouth b) SRF/RES dosing system

4.2.2 Coal milling equipment

A coal preparation rig for drying, milling and mixing allows the usage of different coal ranks. By using different aerodynamic classifiers, a wide range of particle sizes is possible. Coal feed to the main burner is realised by a pressurized dosing unit in order to achieve a nearly non-pulsating flame. The feed rate can be controlled by the weight loss of the discontinuously filled fluidized bed hopper.

4.3 Electrically heated BFB/CFB reactor (ELWIRA)

Main element of the reactor is a stainless steel tube with a diameter of 108 mm in the bed section and 135 mm in the freeboard section. The reactor has an overall height of 3000 mm. A schematic overview is given in the following diagram:

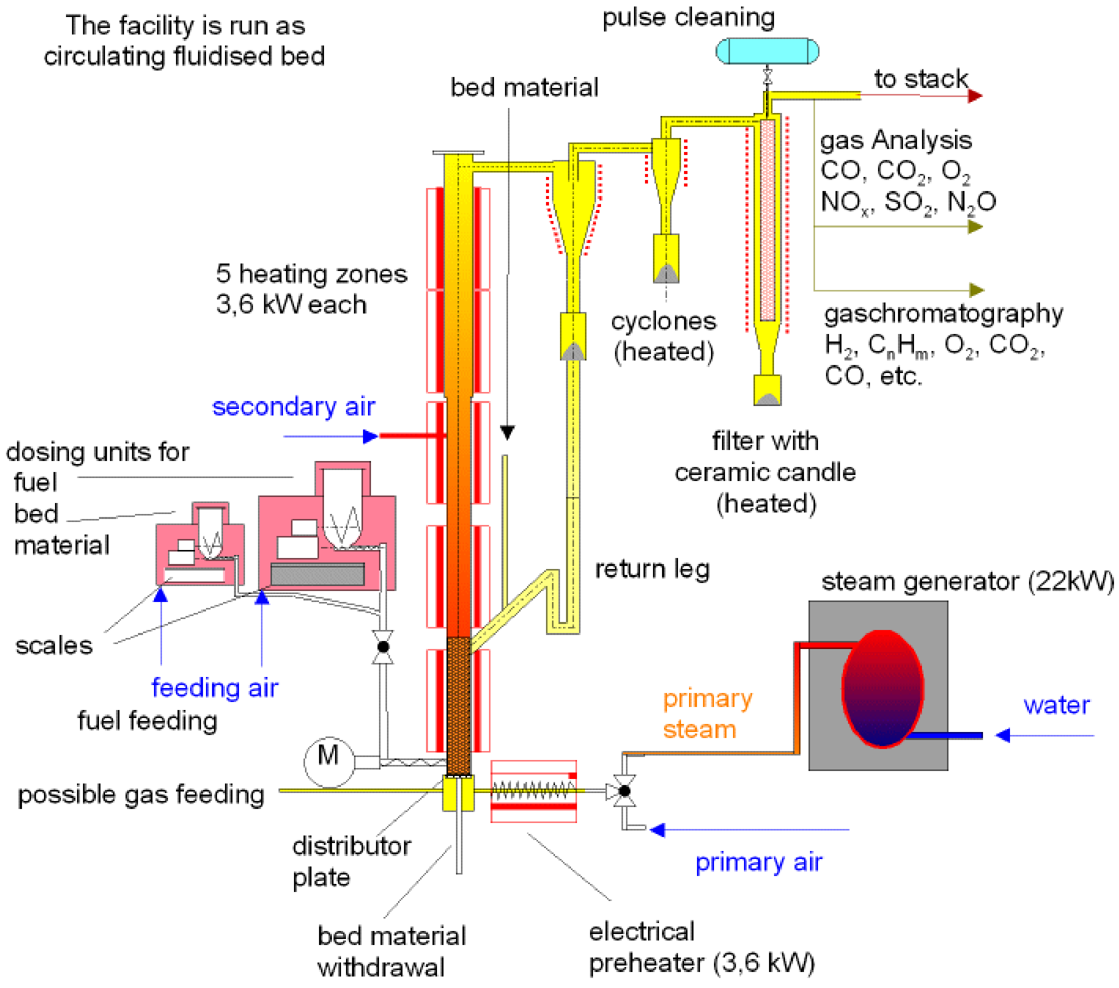

Fig. 4-5: Flow diagram of BFB/CFB reactor ELWIRA

Along its axis the reactor is equipped with different locations for secondary air injection. The reactor has five separately controlled electrical heating zones with approx. 3.6 kW_{el} each. The maximum temperature is 1000°C. In case of CFB operation, the recirculation loop can be heated up to 600°C. Primary air can be pre-heated as well (up to 600°C). The test facility can be operated with coal, biomass, or waste derived fuels. The fluidisation velocity ranges between 0.2-1.0

m/s in bubbling mode (BFB) and 1.0-2.0 m/s in circulating mode (CFB). Ash removal is realised in a two-stage process: A cyclone operated at 700°C and a ceramic candle filter at 150°C. The bed material consists of quartz sand, whereas different particle sizes have to be used for either BFB (200-1000 µm) or CFB (60-200 µm) mode. A coal mass stream between 0.2 and 2 kg/h can be fed, corresponding to a thermal input of 2 to 20 kW. During measurement, flue gases are extracted at the top of the recirculation loop with a cyclone and a bag filter. Standard emissions analysed are O_2, CO_2, CO, SO_x and NO_x. To characterise the volatilisation, char and emission formation, char and fly ash particle samples can be collected at the end of the test runs from the bed material, from the cyclone and the bag filter.

4.4 Artificial waste flue gas generation

As described in chapter 3.1.3, the experiments performed for the UPSWING process required the injection of waste flue gases into the coal combustion process. It was decided to use a gas mixing station, which allows the controlled mixing of gases and the evaporation of different solutions.

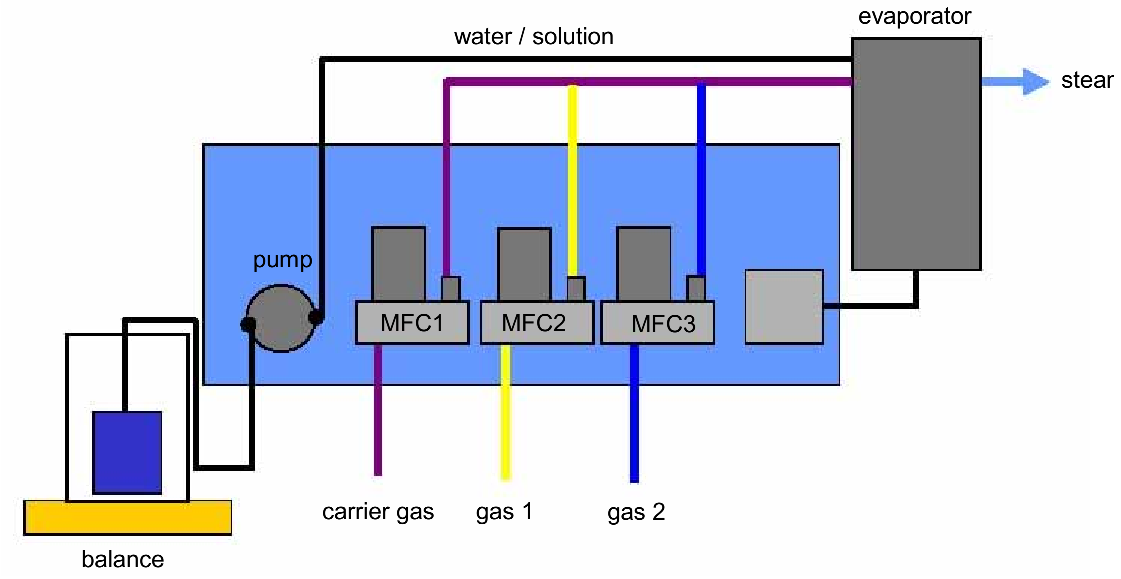

Fig. 4-6: Flow diagram of gas mixing and evaporator unit

The main components of this flue gas generation system are two digitally controlled gas-mixing devices. Up to four different gases can be connected and mixed with the system. The gas flows are controlled by MFCs. Each gas-mixing device is equipped with a vaporiser unit that allows the simple moistening of an artificial flue gas as well as the evaporation of almost every solved component/pollutant such as HCl / HCl(aq), SO_2 / H_2SO_3(aq), NO_2 / HNO_3(aq) and Hg / $HgCl_2$(aq).

4.5 Mercury measurements

4.5.1 Continuous mercury measurements (CMM)

A SEMTECH Hg 2010 continuous mercury monitor (CMM) was used to continuously detect gaseous elemental mercury. A schematic sketch of the CMM is shown in Fig. 4-7. It consists of a wet chemical reduction unit with $SnCl_2$ in a diluted HCl-solution as reducing agent and an analysing unit based on cold-vapour atomic absorption at 253.7 nm. The analysing unit consists of the detection unit, including measurement cell, UV-source and UV-detectors, and a valve to perform the auto-zero procedure, as well as a control unit, and a unit for data preparation, processing, and acquisition. The UV-source (mercury-lamp) emits a light wave at 253.7 nm that is specifically absorbed by elemental mercury. To avoid interferences with other gas components the detection unit uses the method of differential absorption based on the Zeeman-effect for background correction.

To distinguish between the gaseous mercury species $Hg^0(g)$ and the oxidised forms of gaseous Hg^{2+}, mainly present in the chloride form $HgCl_2(g)$, the share of elemental mercury is detected by discontinuously application of an adsorption resin (DOWEX®). DOWEX® is a polystyrol resin with tri-methyl ammonium as functional group. The functional group is completely converted to the chloride form by treatment with HCl. $HgCl_2(g)$ reacts with chloride of the resin by complexing to $[HgCl_3]^-$ or $[HgCl_4]^{2-}$ and adsorption as anion.

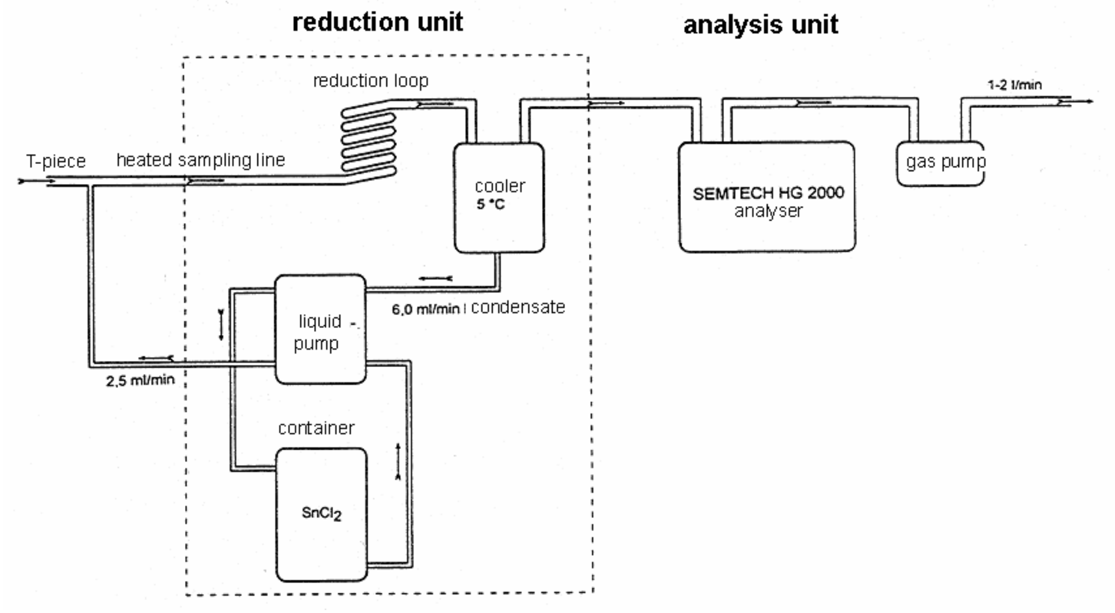

Fig. 4-7: Continuous mercury monitor (CMM)

The efficiency of this selective adsorption of $HgCl_2(g)$ by the adsorption resin ranges from 97 to almost 100 % within a temperature range between 110°C and 140°C. More detailed information about sampling efficiency as well as preparation of the absorption resin is given in [24].

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[24] M. METZGER, H. BRAUN: "In-situ mercury speciation in flue gas by liquid and solid sorption systems"; Chemosphere 16 No.4, pp. 821-832, (1987)

4.5.2 Discontinuous mercury measurements

An alternative method to the above-described CMM is the discontinuous mercury measurement, which was applied during the co-combustion trials. Fig. 4-8 shows the employed mercury measurement setup. The figure shows, that the flue gas is extracted from the flue gas channel via a quartz filter which is located at the end of a glass probe. The glass probe is followed by a two-stage absorption unit, which consists of two glass pipes. The first one is filled with the absorber resin DOWEX® and the second contains iodised charcoal. DOWEX® selectively absorbs the chloride form of gaseous mercury $HgCl_2$(g). The iodised char coal absorbs the other gaseous mercury species which is mainly elemental mercury Hg^0(g). The two absorption pipes are heated to about 100 - 120°C. Then, the water vapour is separated by two wash bottles, where the first is filled with water to cool the gas down and condense the water vapour. The second wash bottle is filled with silica gel in order to ensure that the gas is dry when it enters the pump and the gas meter. The gas pump is adjusted to suck a constant volume flow through the heated sampling train and the wash bottles into the gas meter. The oxygen content is measured downstream the gas meter.

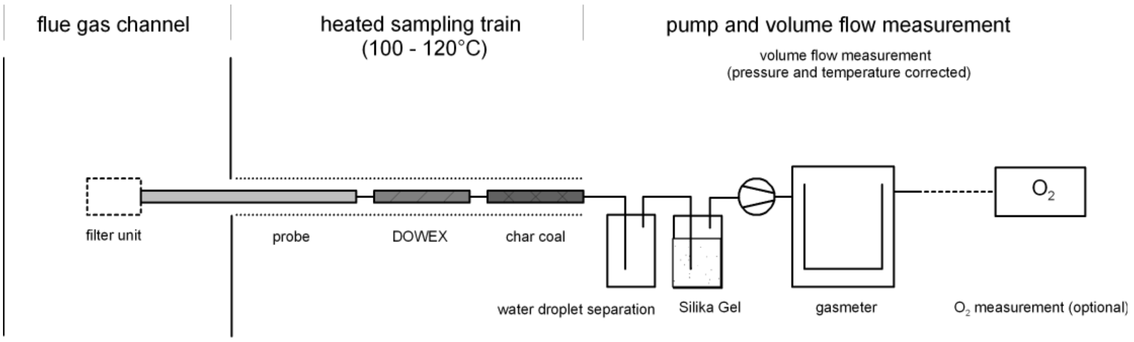

Fig. 4-8: Discontinuous mercury sampling set-up

After the actual measurement the mercury concentrations in the resin and the charcoal are analysed in the laboratory. Together with the other measured parameters, the gas phase mercury concentration is calculated.

4.6 Hydrogen chlorine measurements

Hydrogen chloride was measured following German VDI guideline 3480 [25]. A flue gas sample of 15 l/min is sucked through a heated sampling probe and two adsorption vessels in line, which are filled with distilled water. A heated quartz wool filter is installed between the sampling probe and the absorption vessels to remove particulate matter from the gas. Ion chromatography (IC) was used for analysis of the aqueous samples, detecting anions such as fluoride, chloride, nitrite, nitrate, and sulphate in the range of ppm. IC is a specific form of liquid chromatography that uses ion-exchange resins to separate atomic or molecular ions based on their interaction with the resin.

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[25] German VDI guideline 3480 “Measurement of the hydrogen chloride concentration in waste gases with a low content of particulate chloride”, VDI-Kommision Reinhaltung der Luft, VDI-Verlag GmbH

5 Experimental investigations and discussion

Subject of the previous chapter was a detailed review of the experimental facilities utilised within the scope of this thesis. The next chapter will be dedicated to the experimental results acquired during the trials. Following the priorities for both investigated processes defined in chapter 3, the impact of both waste flue gas integration and direct SRF co-combustion on the power plant process will be investigated in detail.

5.1 Nitrogen oxides

Nitrogen oxides (NO/NO_2 or NO_x) are formed during the combustion process by oxidation of fuel nitrogen or by oxidation of air nitrogen at high temperatures. Three different formation paths are known, of which the oxidation of fuel nitrogen is the most important for coal fired plants (with dry ash removal) and waste incineration processes [26]. Nitrogen oxides are known as toxic substances for human and animal life. Furthermore, they are of environmental relevance when it comes to issues like smog, photo-chemical formation of near-ground ozone, acid rain, or their role in global warming processes [27].

Back in the 1980s, nitrogen oxides became a major issue in public awareness. As a consequence, NO_x reduction measures were commonly introduced in German large-scale coal combustion plants during the late 80's and early 90's along with stricter environmental regulations coming into force, e.g. 13. BImSchV [28] in the year 1983. NO_x reduction measures are usually distinguished between primary and secondary measures:

- Primary measures include all measures in the furnace such as flue gas recirculation, furnace air/fuel staging, or advanced LowNOx burner concepts.
- Secondary measures include SNCR or SCR reactors in high- or low-dust setting (in front or behind the dust removal system).

Todays NO_x limits are restricted to 200 mg/m³ for coal-fired boilers larger than 300 MW_{el} in mono- and co-combustion operation according to the Large Power Plants Directive 2001/80/EC. This limit of 200 mg/m³ is also valid for waste incineration processes according to the Waste Incineration Directive 2000/76/EC.

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[26] J. ZELKOWSKI: „Kohleverbrennung: Brennstoff, Physik und Theorie“, Band 8 der Fachbuchreihe Kraftwerkstechnik, VGB Kraftwerkstechnik, Essen, 1. Auflage, (1986)

[27] G. BAUMBACH: „Luftreinhaltung“, Springer Verlag, ISBN 3-540-56823-9, 3. Auflage, (1993)

[28] Dreizehnte Verordnung zur Durchführung des Bundes-Immissionsschutzgesetzes, Verordnung über Großfeuerungsanlagen, 13 BImSchV, BGBl. I, S. 719, (1983), BGBl. I, S. 632, (2000)

The primary source of NO_x during waste combustion is waste-bound nitrogen as a consequence of the moderate combustion temperatures. Modern waste incinerators are equipped with primary NO_x reduction measures in the furnace, although the scale of these measures is limited as the combustion process on the grate may suffer. Considering the primary objectives of waste incineration, waste inertisation (or complete burnout) and low concentrations of unburned carbon and other products of incomplete combustion (PIC) in the gas phase [29], the use of secondary NO_x reduction measures is inevitable in order to keep the regulatory limit of 200 mg/m³. Due to fluctuations in the waste nitrogen content, raw flue gas concentrations in the range of 100-500 mg/m³ can be expected [17].

The UPSWING concept foresees no reduction or abatement of nitrogen oxides in the (partial) flue gas treatment system of the waste incinerator. It is one of the main ideas of the concept to reduce these emissions in the coal boiler, favourably in the reduction zone of the combustor. An investigation of the impact of flue gas integration on overall NO_x emission and towards the NO_x reduction potential in the process is therefore necessary. The main objectives of the investigations performed within the scope of this work can be summarised as follows:

- What are the general consequences of waste flue gas injection on process NO_x emissions? What emission levels can be expected?
- What is the influence of the injection location and which injection concept should be favoured in terms of low NO_x emissions?
- What is the influence of the coal combusted in the power plant process? Are there differences between individual coals?

The experiments were performed based on three different coals at the BTS combustion reactor (refer to chapter 4.1). Artificial waste incinerator flue gases were generated and injected at different locations as outlined in chapter 3.1.3. The results of these experiments were compared with the respective reference case (100% coal combustion). Furthermore, the evaluation of a favourable injection concept was - based on the experimental results obtained - investigated by numerical simulation. The results of this more theoretical approach are summarised in chapter 6. In terms of direct SRF co-combustion, experiments were performed at the KSVA combustion facility (refer to chapter 4.2). The emission behaviour of the co-combustion process was compared to the reference case corresponding to the experiments performed for the UPSWING process.

5.1.1 Emission behaviour of the UPSWING process

As mentioned before, the partial flue gas cleaning concept is not designed to reduce NO_x in the raw waste flue gases. Therefore, it is important that the additional pollutants are reduced in the coal boiler. This is of special relevance for brown coal fired boilers, which are usually not equipped with secondary NO_x

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[29] International ash working group: A.J. CHANDLER, T.T. EIGHMY, J. HARTLEN, O. HJELMAR, D.S. KOSSON, S.E. SAWELL, H.A. van der SLOOT, J. VEHLOW: “Municipal solid waste incinerator residues”, Elsevier, Amsterdam

reduction measures. Maintaining the environmental standards is therefore crucial for successful implementation of the UPSWING concept. To investigate this important topic, artificial waste flue gases have been injected during coal combustion operation into the BTS combustion reactor. A basic composition of the dry waste flue gases consisting of 80% N_2, 10% O_2, and 10% CO_2 as an average waste gas composition was used during the trials. During the trials, the NO_x content of the injected waste flue gas was increased from 0 to 400 ppm (approx. 800 mg/m³) to cover the usual range of NO_x in waste flue gases (see chapter 3.1.2) and to investigate the fate of this additional pollutant load. The influence of possible side reactions of intermediate species was not investigated in the context of this parameter study.

The impact of waste flue gas integration on the coal combustion process was investigated for three different coals, a Colombian bituminous coal (B/1), a Polish lignite (L/1), and a Czech hard brown coal (L/2). A characterisation of the fuels can be found in chapter 3.1.3. The waste flue gas ratio was adjusted to 10% in relation to the dry flue gas volume produced by combustion of the investigated coals. The relevant combustion settings for the coals B/1, L/1, and L/2 are summarised in the following table:

Table 5-1: Basic combustion settings

Parameter / Coal	Coal B/1	Coal L/1	Coal L/2
Coal mass flow	1.00 kg/h	1.00 kg/h	1.00 kg/h
Reactor temperature	1100°C	1100°C	1100°C
Combustion air (λ=1.20)	8.46 m³/h	5.37 m³/h	7.15 m³/h
Flue gas coal (dry)	8.08 m³/h	5.16 m³/h	6.86 m³/h
Flue gas coal (wet)	8.86 m³/h	5.71 m³/h	7.54 m³/h
10% MSWI flue gas (dry)	0.81 m³/h	0.52 m³/h	0.69 m³/h
Additional NO load	0, 200, 400 ppm	0, 200, 400 ppm	0, 200, 400 ppm
Additional H_2O load	0.38 kg/m³	0.38 kg/m³	0.38 kg/m³

Considering the oxygen content of 10 vol.-% in the waste flue gases, two different operational cases have been investigated: The additional injection without altering the coal burner air settings (Case A) and the partly substitution of secondary burner air by the waste flue gases (Case B) in order to achieve constant O_2 levels at the furnace exit. The combustion settings for both investigated cases are summarised in the following table:

Table 5-2: Combustion air and flue gas amount for additional flue gas injection (Case A)

Case A (additional)	Coal B/1	Coal L/1	Coal L/2
Combustion air	8.46 m³/h	5.37 m³/h	7.15 m³/h
Flue gas total (dry)	8.89 m³/h	5.67 m³/h	7.55 m³/h
Flue gas total (wet)	9.86 m³/h	6.34 m³/h	8.39 m³/h
O_2 level at furnace exit	4.1 vol.-%	4.1 vol.-%	4.1 vol.-%

Table 5-3: Combustion air and flue gas amount for substitution of combustion air (Case B)

Case B (substitution)	Coal B/1	Coal L/1	Coal L/2
Combustion air	8.06 m^3/h	5.17 m^3/h	6.81 m^3/h
Substituted combustion air	0.40 m^3/h	0.20 m^3/h	0.34 m^3/h
Flue gas total (dry)	8.49 m^3/h	5.56 m^3/h	7.22 m^3/h
Flue gas total (wet)	9.46 m^3/h	6.24 m^3/h	8.05 m^3/h
O_2 level at furnace exit	3.5 vol.-%	3.5 vol.-%	3.5 vol.-%

Considering the overall stochiometry in the furnace, it should be clear that the additional injection of the waste flue gas - case A - increases the total available oxygen. Contrary to this, the total available oxygen in the furnace is constant for case B as the secondary air is substituted by the waste flue gases. During the trials, two basic injection locations have been investigated, viz. the flue gas injection into the secondary air inlet of the burner (option 1) and the injection via a lateral access port in 0.9m distance from the burner (option 2). The relevance of both injection locations in terms of large-scale application has already been discussed in chapter 3.1.3. Considering both injection locations as well as both operational cases A/B, it is obvious that the stochiometry at the burner is changed. This is of special relevance for the experiments during staged and unstaged coal combustion, which will be discussed for the investigated coals.

a) Results for the Colombian bituminous coal B/1

Table 5-4 summarises the measured NO_x emissions for the Colombian bituminous coal (B/1) for the investigated settings.

Table 5-4: Measured NO_x emissions in mg/m^3_N tr. 6% for the bituminous coal B/1

Mode	A.1		A.2		B.1		B.2	
Conditions	unstaged	staged	unstaged	staged	unstaged	staged	unstaged	staged
Baseline	1129	290	1129	290	1129	290	1129	290
0 ppm	1183	372	1157	219	1068	294	1119	202
200 ppm	1191	379	1203	217	1045	290	1160	196
400 ppm	1199	381	1214	225	1076	292	1149	205

In this context, “A.1” refers to additional waste flue gas injection via the secondary air nozzles of the burner, “A.2” to additional injection via the lateral access port at 0.9m, “B.1” to substitution of secondary air and injection via the secondary air nozzle, and “B.2” to substitution of secondary air and injection via the lateral access port at 0.9m. The combustion conditions are either unstaged or staged, while during staged injection a part of the combustion air was injected as burnout air at 1.4 m distance from the burner (OFA). “Baseline” refers to the results of 100% coal operation. The ratio of waste flue gas in relation to the flue gas from coal is 10%, and the NO_x concentration of the waste flue gases is 0, 200, and 400 ppm.

Prior to the discussion of the results a further step is required. To ensure a later process comparison to SRF co-combustion on a common basis, the introduction of process emission factors is necessary. The process emission factor relates the measured pollutant concentrations to the total heat input of the process and can be calculated according to equation 5.1:

$$e_i = \frac{c_i^{FG} \cdot \dot{V}_{FG}}{\dot{Q}_{input}} \qquad \text{g/MWh} \qquad (5.1)$$

The resulting emission factor is expressed in g/MWh. The corresponding heat input for the process derives from the coal and the combusted municipal solid waste:

$$\dot{Q}_{input} = \dot{m}_{coal} \cdot NCV_{coal} + \dot{Q}_{MSW} \qquad \text{MWh} \qquad (5.2a)$$

Based on an average NCV of waste of 8680 kJ/kg the 10% flue gas ratio was correlated to a theoretical heat input from MSW of 0.477 kWh for the experiments with the bituminous coal. Table 5-5 summarises the calculated process emissions:

Table 5-5: Specific NO_x process emissions in g/MWh for the bituminous coal B/1

Mode	A.1		A.2		B.1		B.2	
Conditions	unstaged	staged	unstaged	staged	unstaged	staged	unstaged	staged
Baseline	1220	313	1220	313	1220	313	1220	313
0 ppm	1321	415	1292	245	1138	313	1193	215
200 ppm	1330	423	1343	242	1114	309	1236	209
400 ppm	1339	425	1355	251	1147	311	1224	218

For better understanding of the entire approach, the heat input during co-combustion shall be mentioned as well and is calculated according to equation 5.2b:

$$\dot{Q}_{input} = \dot{m}_{coal} \cdot NCV_{coal} + \dot{m}_{SRF} \cdot NCV_{SRF} \qquad \text{MWh} \qquad (5.2b)$$

A comparison of process specific emissions is possible by the introduction of a specific emissions factor ef_i (%), calculated as the ratio of the specific process emission and specific process emission during baseline operation ($e_{i,0}$):

$$ef_i = \frac{e_i}{e_{i,0}} \qquad \% \qquad (5.3)$$

Table 5-6 shows the calculated process emission factors for the bituminous coal B/1. Following the definition of the factor, the baseline cases are always 100%.

Table 5-6: Specific NO_x process emission factors in % for the bituminous coal B/1

Mode	A.1		A.2		B.1		B.2	
Conditions	unstaged	staged	unstaged	staged	unstaged	staged	unstaged	staged
Baseline	100%	100%	100%	100%	100%	100%	100%	100%
0 ppm	108%	133%	106%	78%	93%	100%	98%	69%
200 ppm	109%	135%	110%	77%	91%	99%	101%	67%
400 ppm	110%	136%	111%	80%	94%	99%	100%	70%

Fig. 5-1 shows the process emission factors for the trials with the bituminous coal B/1 for unstaged combustion conditions. Results larger then 100% indicate higher specific emissions compared to the baseline case, which indicate increased process emissions of the entire process.

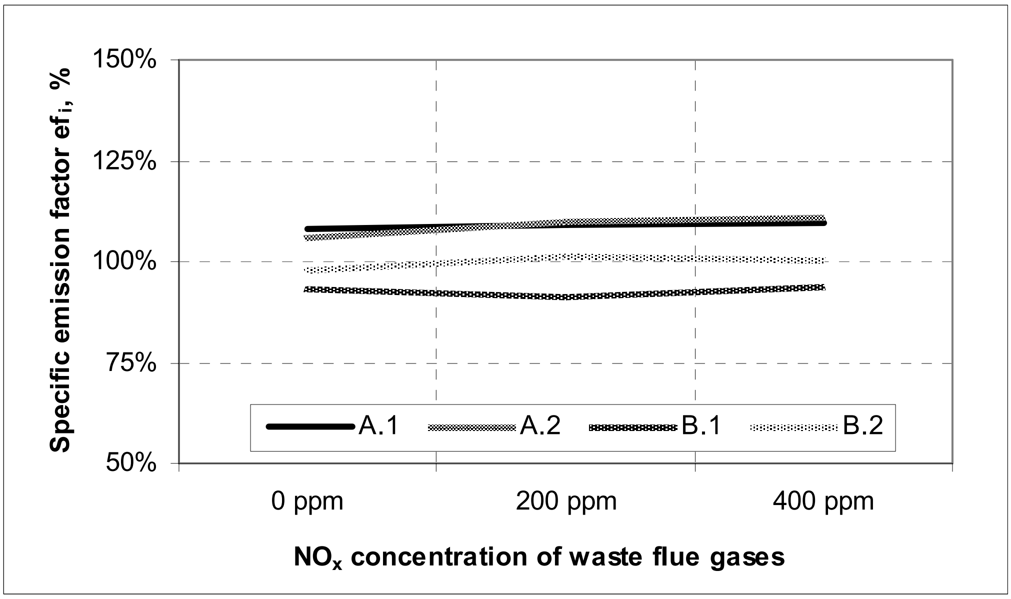


Fig. 5-1: Process emission factors for the bituminous coal B/1, unstaged combustion conditions

For the bituminous coal, the specific process emissions range between 90 and 110% for unstaged combustion. Both cases A.1 and A.2 show slightly increased process emissions, presumably due to increased oxygen content in the furnace. Nevertheless, no major influence of the respective injection location can be determined. Case B.2 shows almost the same levels as baseline coal combustion despite the fact that the burner stochiometry is lower in this case. Consequently, lower emission would be expected. Instead, lower process emissions were found for case B.1, but this result can only be explained by a reduction of the flame temperature as a consequence of waste flue gas injection.

Fig. 5-2 shows the process emission factors for the trials with the bituminous coal B/1 for staged combustion conditions. In case of stage combustion conditions, the deviations between the different operational settings are much larger compared to unstaged combustion. The specific emission factors range between 70 and 135%, indicating that NO_x formation and reduction processes will react quite sensitive upon the injection of waste flue gases. Furthermore, and as a key statement, there is apparently no significant influence of the additional NO_x load. Considering the investigated cases, case A.1 shows increased emissions compared to baseline, while case B.1 is almost comparable. For both cases, the results can be explained with an altered burner stochiometry. Considering the injection via the lateral access port at 0.9m (cases A.2 and B.2) shows that waste flue gas injection would allow even lower process emissions than baseline coal combustion. The minor difference between both cases indicates that NO_x formation/reduction processes are almost completed at the injection location.

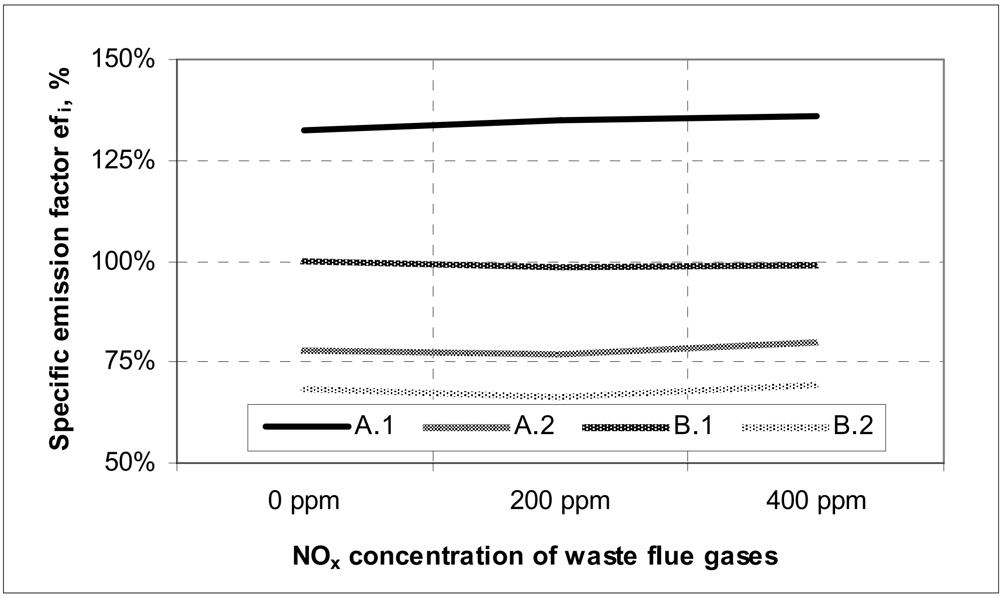


Fig. 5-2: Process emission factors for the bituminous coal B/1, staged combustion conditions

b) Results for the Polish lignite L/1

The discussion for the experiments with the Polish lignite will follow the same pattern as described for the bituminous coal discussed above. Table 5-7 summarises the measured NO_x emissions for the lignite:

Table 5-7: Measured NO_x emissions in mg/m³$_N$ tr. 6% for the Polish lignite L/1

Mode	A.1		A.2		B.1		B.2	
Conditions	unstaged	staged	unstaged	staged	unstaged	staged	unstaged	staged
Baseline	1062	211	1062	211	1062	211	1062	224
0 ppm	1074	340	1084	233	1069	209	1102	224
200 ppm	1081	411	1100	241	1092	208	1112	219
400 ppm	1078	477	1123	243	1097	205	1110	214

Based on an average NCV of waste of 8680 kJ/kg the 10% flue gas ratio was correlated to a theoretical heat input from MSW of 0.314 kWh for the experiments with the Polish lignite. Table 5-8 summarises the calculated process emissions:

Table 5-8: Specific NO_x process emissions in g/MWh for the Polish lignite L/1

Mode	A.1		A.2		B.1		B.2	
Conditions	unstaged	staged	unstaged	staged	unstaged	staged	unstaged	staged
Baseline	1267	252	1267	252	1267	252	1267	267
0 ppm	1315	416	1327	285	1249	244	1288	262
200 ppm	1323	503	1347	295	1276	243	1300	256
400 ppm	1320	584	1375	298	1282	240	1297	250

The results show a significant increase of the specific process emissions for case A.1 during staged combustion. Taking the other results as well as the results for both other coals into account, the results obviously do not fit. The reason for this deviation is not clear, but potentially related to operational problems (e.g. coal feeding) during the trials. The calculated emission factors according to equation 5.3 are summarised in the following table:

Table 5-9: Specific NO_x process emission factors in % for the Polish Lignite L/1

Mode	A.1		A.2		B.1		B.2	
Conditions	unstaged	staged	unstaged	staged	unstaged	staged	unstaged	staged
Baseline	100%	100%	100%	100%	100%	100%	100%	100%
0 ppm	104%	165%	105%	113%	99%	97%	102%	98%
200 ppm	104%	200%	106%	117%	101%	97%	103%	96%
400 ppm	104%	232%	109%	118%	101%	95%	102%	94%

Fig. 5-3 shows the process emissions factors for the trials with the Polish Lignite L/1 for unstaged combustion conditions.

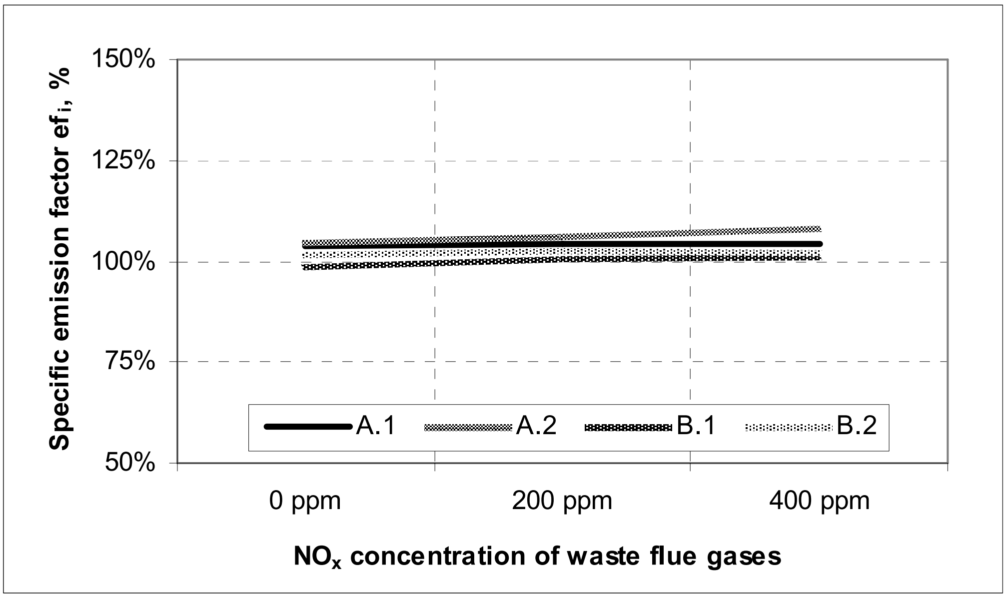


Fig. 5-3: Process emission factors for the Polish Lignite L/1, unstaged combustion conditions

Contrary to the results for the bituminous coal, the results show increased process emissions for almost each investigated case. The results for additional injection (cases A.1 and A.2) are slightly higher compared to the substitution of secondary air (cases B.1 and B.2), although the minor differences make an interpretation of the results difficult. A slight dependence of the results in terms of the waste flue gas NO_x concentration can be stated. Fig. 5-4 shows the process emissions factors for the trials with the Polish lignite L/1 for staged combustion conditions.

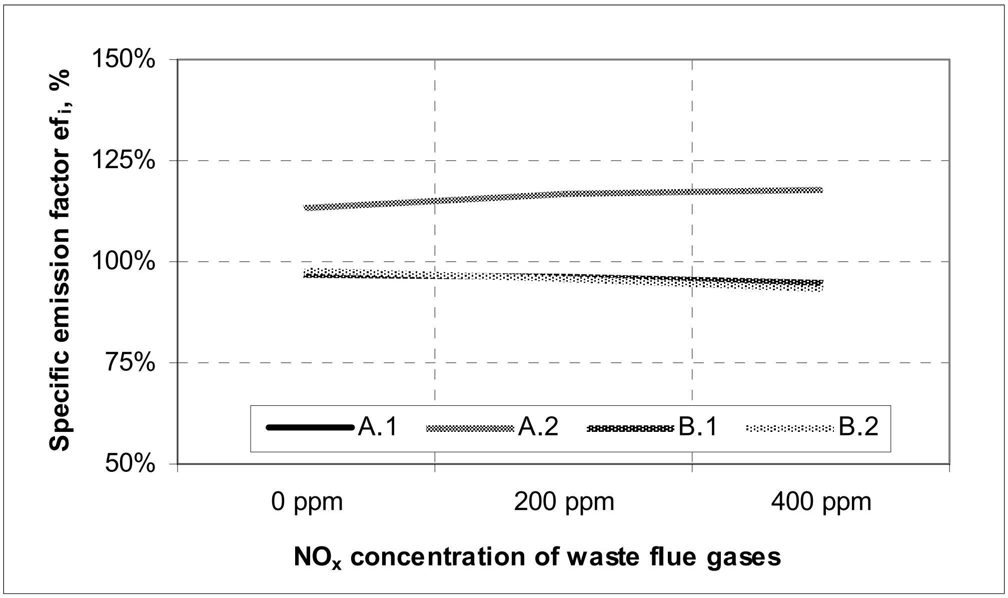


Fig. 5-4: Process emission factors for the Polish lignite L/1, staged combustion conditions

In case of staged combustion conditions, the results for the lignite show a clear influence between additional waste flue gas injection (case A.2) and the substitution of secondary air (cases B.1 and B.2). The specific process emissions are increased in case of A.2. Considering B.1 and B.2, there is obviously no significant influence of the injection location, indicating a much longer NO_x formation/reduction zone for the lignite compared to the bituminous coal. Despite of this, the results show that comparable process emissions are generally possible, although the stochiometry has to be maintained. Comparable to the results of the bituminous coal, there is obviously no major influence of the additional NO_x load during staged combustion conditions.

b) Results for the Czech hard brown coal L/2

The last investigated coal is the Czech hard brown coal L/2. Table 5-10 summarises the measured NO_x emissions for the investigated settings.

Table 5-10: Measured NO_x emissions in mg/m^3_N tr. 6% for the Czech hard brown coal L/2

Mode	A.1		A.2		B.1		B.2	
Conditions	unstaged	staged	unstaged	staged	unstaged	staged	unstaged	staged
Baseline	1336	230	1336	235	1336	230	1336	230
0 ppm	1271	285	1264	207	1262	234	1176	185
200 ppm	1276	287	1288	206	1278	230	1187	184
400 ppm	1297	293	1293	209	1280	234	1204	188

Based on an average NCV of waste of 8680 kJ/kg the 10% flue gas ratio was correlated to a theoretical heat input from MSW of 0.386 kWh for the experiments with the Czech hard brown coal. Table 5-11 summarises the calculated process emissions:

Table 5-11: Specific NO_x process emissions in g/MWh for the Czech hard brown coal L/2

Mode	A.1		A.2		B.1		B.2	
Conditions	unstaged	staged	unstaged	staged	unstaged	staged	unstaged	staged
Baseline	1803	310	1803	317	1803	310	1803	310
0 ppm	1762	395	1752	287	1670	310	1556	245
200 ppm	1768	398	1785	286	1691	304	1570	243
400 ppm	1798	406	1792	290	1693	310	1593	249

The calculated emission factors according to equation 5.3 are summarised in the following table:

Table 5-12: Specific NO_x process emission factors in % for the Czech hard brown coal L/2

Mode	A.1		A.2		B.1		B.2	
Conditions	unstaged	staged	unstaged	staged	unstaged	staged	unstaged	staged
Baseline	100%	100%	100%	100%	100%	100%	100%	100%
0 ppm	98%	127%	97%	90%	93%	100%	86%	79%
200 ppm	98%	128%	99%	90%	94%	98%	87%	78%
400 ppm	100%	131%	99%	91%	94%	100%	88%	80%

Fig. 5-5 shows the process emissions factors for the trials with the Czech hard brown coal L/2 for unstaged combustion conditions.

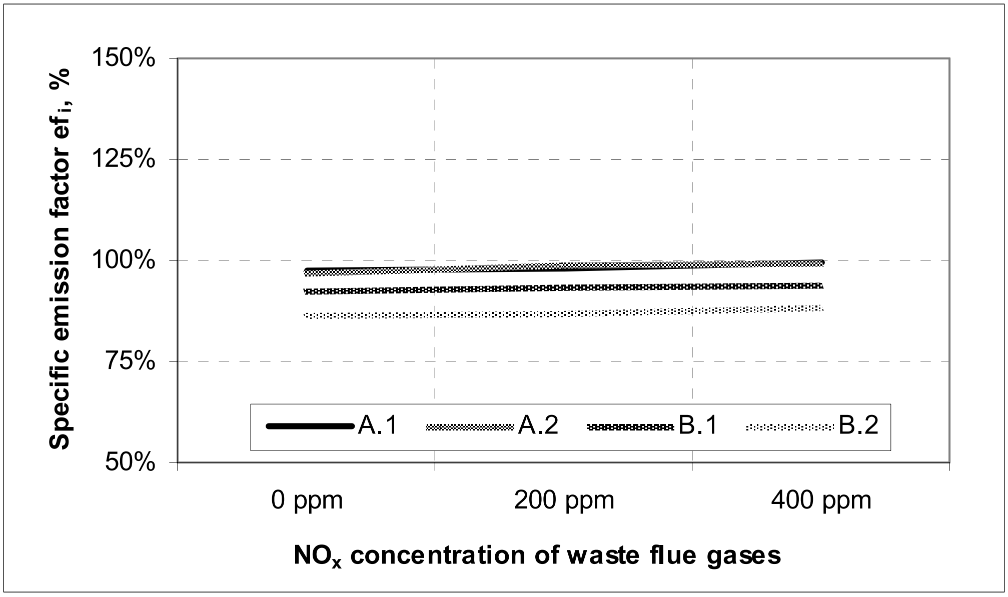


Fig. 5-5: Process emission factors for Czech hard brown coal L/2, unstaged combustion conditions

Contrary to the other coals, the results indicate lower specific process emissions for any investigated case. Considering the cases A.1 and A.2, no significant difference could be observed, and the resulting process emissions are close to baseline coal operation. Nevertheless, no major influence of the respective injection location was found during unstaged combustion conditions. Considering the cases B.1 and B.2, lower process emissions could be observed for the injection via the lateral access port at 0.9m. This result can be explained with the lower burner stochiometry in case of B.2. Fig. 5-6 shows the process emissions factors for the trials with the Czech hard brown coal L/2 for staged combustion conditions.

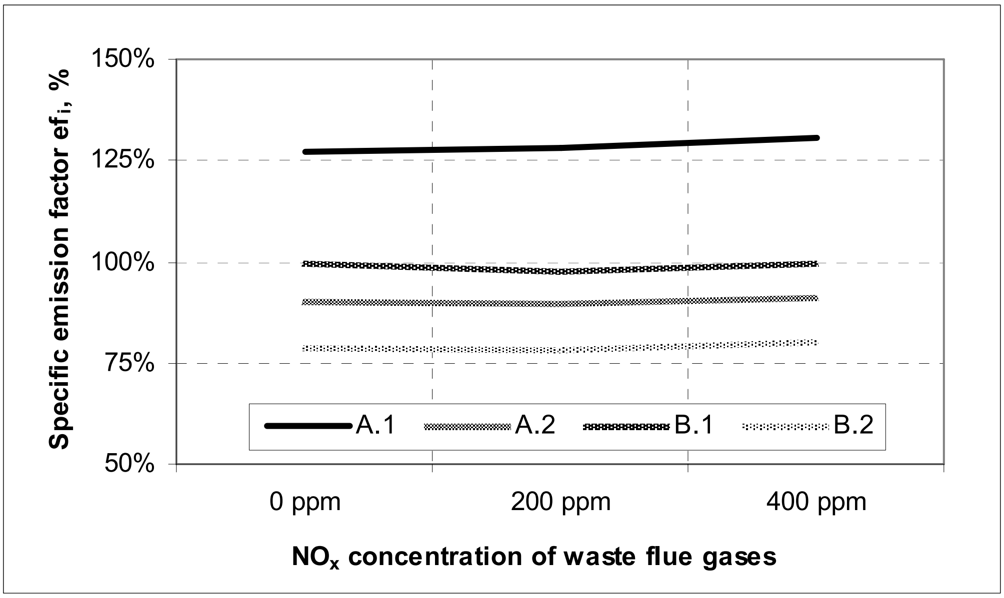


Fig. 5-6: Process emission factors for Czech hard brown coal L/2, staged combustion conditions

Considering the investigated cases, case A.1 shows significant higher emissions compared to baseline coal combustion. Case B.1 is almost comparable to baseline. For this specific coal, the injection via the lateral injection at 0.9m leads to favourable results. Generally, the results for the Czech hard brown can be well correlated with the results for the bituminous coal. As the results for the Polish lignite L/1 differ significantly, the influence of waste flue gas injection on process emissions seems to depend strongly on the specific coal properties.

5.1.2 Process emissions during SRF co-combustion

The characterisation of the process emission behaviour during SRF co-combustion should be discussed based on the experiences elaborated at the semi-technical combustion facility KSVA. A description of the test facility can be found in chapter 4.2. Experiments were performed using two German bituminous coals (B/2 and B/3) and one SRF quality (SRF/PRR) in two different thermal shares (5 and 10%). The bituminous coal B/2 has a nitrogen content of 1.7 wt.-%, the bituminous coal B/3 of 1.5 wt.-%. The nitrogen content of the SRF material - 0.2 wt.-% - is significantly lower. Therefore, lower emissions can be expected during SRF co-combustion. An assessment of the reduction potential is therefore the primary task. To ensure comparable conditions during the trials, the power input into the test facility was set to 300kW. The respective mass flows of both coal and SRF are summarised in Table 5-13 and Table 5-14:

Table 5-13: Combustion settings bituminous coal B/2 – SRF/PRR

Coal, kg/h	P, kW	SRF share	SRF, kW	SRF, g/h	Ash, kg/h	P, kW
35.00	299.79	0.0%	0.00	0	2.87	299.79
33.25	284.80	5.0%	14.99	2367	2.93	299.79
31.50	269.81	10.0%	29.98	4734	2.99	299.79

Table 5-14: Combustion settings bituminous coal B/3 – SRF/PRR

Coal, kg/h	P, kW	SRF share	SRF, kW	SRF, g/h	Ash, kg/h	P, kW
33.50	301.85	0.0%	0.00	0	1.81	301.85
31.83	286.76	5.0%	15.09	2383	1.93	301.85
30.15	271.67	10.0%	30.19	4767	2.04	301.85

Comparable to the experiments discussed in the previous chapter, specific process emission factors were calculated for the co-combustion process according to equation 5.1 through 5.3. The results for both coals are summarised in Table 5-15 and Table 5-16:

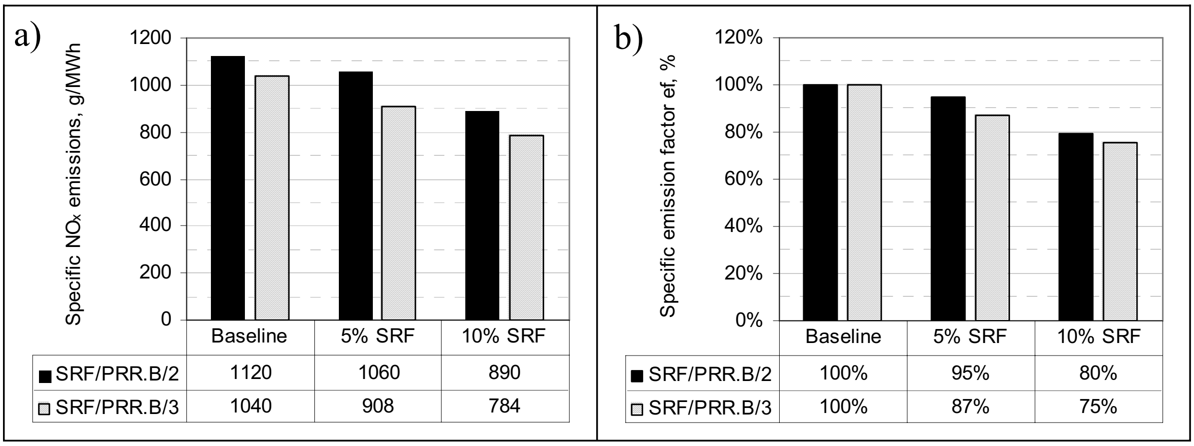
Table 5-15: Emission factor for bituminous coal B/2 – SRF/PRR

SRF/PRR.B/2	Baseline	$5\%_{th}$	$10\%_{th}$
$\dot{V}$, m³/h	330.7	330.5	330.3
$\dot{Q}$, MJ/h	1079.4	1079.4	1079.4
c_{NOx}, mg/m³	1015	962	808
e_{NOx}, g/MWh	1119.5	1060.4	890.1
ef_{NOx}, %	100.0%	94.7%	79.5%

Table 5-16: Emission factors for bituminous coal B/3 – SRF/PRR

SRF/PRR.B/3	Baseline	$5\%_{th}$	$10\%_{th}$
$\dot{V}$, m³/h	331.1	330.9	330.7
$\dot{Q}$, MJ/h	1086.7	1086.7	1086.7
c_{NOx}, mg/m³	948	828	716
e_{NOx}, g/MWh	1039.8	907.7	784.4
ef_{NOx}, %	100.0%	87.3%	75.4%

The results show a significant decrease of measured NO_x as a consequence of an increased SRF share, which can be well explained with the lower nitrogen content of the specific SRF material with a nitrogen content of 0.2%. As shown in Fig. 5-7, a good consistency between the results for the two investigated coals was found.



	Baseline	5% SRF	10% SRF
■ SRF/PRR.B/2	1120	1060	890
□ SRF/PRR.B/3	1040	908	784

	Baseline	5% SRF	10% SRF
■ SRF/PRR.B/2	100%	95%	80%
□ SRF/PRR.B/3	100%	87%	75%

Fig. 5-7: a) Specific emissions e_{NOx}, g/MWh b) Specific emission factor ef_{NOx}, %

Considering the co-combustion process, the behaviour found follows the expectations as a result of lower nitrogen concentration in the SRF. Furthermore, the lower nitrogen input into the process leads to lower primary pollutant concentrations in the flue gas. The specific process emissions of the bituminous coal B/1 are slightly higher, though this result can be explained with a higher nitrogen content of 1.7 wt.-% compared to 1.5 wt.-% of B/2. Generally, the results show a beneficial aspect of SRF co-combustion in terms of lower process emissions.

5.1.3 Comparison and evaluation of results

Both processes show the potential to reduce NO_x emissions in comparison to baseline (coal) combustion. In case of direct SRF co-combustion, the benefits derive from the lower nitrogen content of the secondary fuel. Lower specific process emissions could be determined for both investigated bituminous coals as a function of the co-combusted share of SRF. Considering the UPSWING process, it became obvious that the location of flue gas injection as well as maintaining the burner stochiometry is crucial in terms of low process emissions. Furthermore, it was found that the reduction of the additional NO_x load of the injected waste flue gases is generally possible. This assumption derives from the fact that no significant influence of an additional pollutant load was determined for any of the investigated coals. Significant differences were found between the investigated coals and their sensitivity upon flue gas injection, especially for the investigated lignite L/1. Generally, the results indicate that the substitution of secondary air by the remaining oxygen of the waste flue gases is the favourable option. With the exception of the lignite L/1, additional injection behind the coal flame (lateral injection) can be recommended for both the investigated bituminous and hard brown coal. Nevertheless it became obvious that minimal process emissions during waste flue gas injection require well controlled process conditions.

5.2 Hydrogen chlorine

Chlorine is brought into the combustion process in inorganic as well as in organic form. A considerable amount is transferred into the gas phase. According to Born, approximately 75% of the initial chlorine input can be found in the flue gas treatment system of MSWI [30]. In technical combustion systems, such as pulverised fuel fired plants or conventional waste incinerators, HCl is the dominant species, evolving when inorcanic chlorides are sulphatised in presence of SO_2. The existence of chlorine in form of Cl_2 is also possible as a result of a direct oxidation of HCl to Cl_2 in presence of metal chlorides ($MeCl_x$) [31]. The Deacon equilibrium plays an important role in this context. Considering direct process emissions, hydrogen chloride is limited to 10 mg/m³ for mono- and co-combustion processes in accordance with the European directive 2000/76/EC, while no limits are defined in the large power plant directive 2001/80/EC. In conventional power plants, particle-bound chlorine is removed with the ash, while volatile chlorine is removed in the flue gas de-sulphurisation system (FGD). Contrary to this, modern MSWI are commonly equipped with a dedicated scrubbing stage to remove chlorine and other acid halogens (refer to chapter 3.1.1).

According to Born, chlorine is the most important species for fire-side corrosion. Relevant chlorine species are gaseous hydrogen chloride (HCl) and elemental chlorine (Cl_2), as well as alkali chlorides found in depositions on heating surfaces. The primary sources for chlorine species in the combustion process are fuel components, but also NaCl in the combustion air of plants which are closely located to coastal areas can increase the chlorine input. The above mentioned chlorine species allow the direct or indirect formation of metal chlorides ($MeCl_x$). According to Reichel [32], both HCl and Cl_2 are capable to form $FeCl_2$ in a direct corrosion attack. The indirect formation of metal chlorides requires an intermediate step to form HCl or elemental Cl_2, e.g. by sulphurisation of alkali salts. In this context, ash-bound chlorine (salts) becomes the primary source of chlorine in ash- or slag layers (fouling) [33]. The condensation of evaporated salts on surfaces is another source of chlorine in surface layers. Both evolving HCl and Cl_2 can directly interact with the base material (e.g. superheater surfaces) as described above.

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[30] M. BORN: „Thermische Abfallbehandlung: Verbrennung, Pyrolyse, Vergasung?”, Thermoprozeß- und Abfalltechnik, Vulkan Verlag, 3. Ausgabe, (1994)

[31] M. HOCQUEL, S. UNDERBERGER, K.R.G. HEIN, J. BOCK: „Behaviour of mercury under different oxidizing conditions“, International conference on air quality, trace elements and particulate matter, Arlington, VA (USA), (2002)

[32] H. H. REICHEL: “Rauchgasseitige Korrosion in fossilbefeuerten deutschen Kraftwerken”, VGB Kraftwerkstechnik 68, Heft 2, S. 186-197, (1988)

[33] L. SINGHEISER: “Hochtemperaturkorrosion in Verdampferanlagen und Maßnahmen zu deren Vermeidung”, VDI Berichte Nr. 773, (1990)

In comparison to (stable) metal oxides, the thermodynamic properties of metal chlorides are completely different. Generally, they show lower melting points and - in a temperature range of 300 to 600°C - high vapour pressures. As a consequence, they can evaporate at relatively modest surface temperatures. Higher material temperatures cause a linear increase of their vapour pressures, resulting in increased corrosion rates. Especially unprotected surfaces are highly endangered by the described mechanisms. This aspect is especially relevant for new installations, for example after replacements during boiler revision, lacking a protective oxide layer. Therefore, a sufficient oxygen level should be maintained during the following start-up procedure. Doing so, the formation of unwanted metal chlorides can be suppressed while the formation of protective oxygen layers is enforced. Furthermore, the destruction of an existing protective oxide layer should be avoided (e.g. by over-temperatures caused by direct flame exposure, boiler trips in water-/steam system, or low-oxygen (reducing) atmospheres) [34].

As a consequence of the above mentioned aspects, the chlorine input into the boiler system should generally be limited, especially during critical situations such as start-ups after revision or in newly-commissioned plants. This approach is valid for both processes discussed in this thesis, UPSWING and direct SRF co-combustion. The partial flue gas treatment of the UPSWING concept includes an acid scrubbing stage in order to remove the major part of chlorine from the raw flue gases. Another advantage is the almost complete ash removal prior to the injection of the partially-cleaned flue gases into the power plant process. An influence on the ash chemistry of the power plant process is therefore limited.

Considering the SRF production process, chlorine reduction is one of its primary targets, although limited by economic and technical aspects as described in chapter 2.2. Comparing the chlorine concentrations found in SRF with coal, they usually show higher concentrations. As a consequence, co-combustion is expected to lead to higher chlorine concentrations in the flue gases. Nevertheless, a higher chlorine concentration in the gas phase does not necessarily mean that a significant increase of corrosion effects takes place, especially if protective oxid layers are already existing and problematic wall atmospheres (low oxygen levels, high temperatures) are avoided. To summarise the previous discussion, chlorine, and especially gaseous chlorine, shall be taken as an indicator for an increased risk of corrosion. This was basically done by a comparison of both processes to baseline coal operation.

5.2.1 Fate of chlorine in the UPSWING process

The fate of additional chlorine was investigated by means of a balance calculation, considering both chlorine from coal combustion and chlorine originating from waste combustion. In chapter 3.1.2, it was shown that the residual chlorine concentration of the waste flue gases ranges between 10 and 40 mg/m³ $HCl_{(g)}$. For the investigated coals, Table 5-17 summarises the theoretical

HCl concentration - based on the elemental fuel analysis - and assuming 100% of the fuel chlorine is released into the gas phase during coal combustion:

Table 5-17: Theoretical HCl concentration of flue gases from coal combustion

Fuel	B/1	L/1	L/2
Cl, $\%_{daf}$	0.05	0.06	0.06
Specific dry flue gas volume, m^3/kg	6.59	4.16	5.66
HCl, mg/m^3	44	71	65

The balance calculation results in a HCl concentration of 44 mg/m^3 for the bituminous coal B/1, of 71 mg/m^3 for the lignite L/1, and of 65 mg/m^3 for the hard brown coal. The given numbers do not include the additional chlorine input of the waste flue gases yet.

To assess the residual HCl concentration of the mixed flue gases from coal and waste combustion, balance calculations were made for the bituminous coal B/1 and the lignite L/1, taking into account the range of 10 to 40 mg/m^3 behind the partial flue gas treatment process. The calculations refer to the thermal share of MSW, replacing up to 25% of the total heat input, as shown in Fig. 5-8:

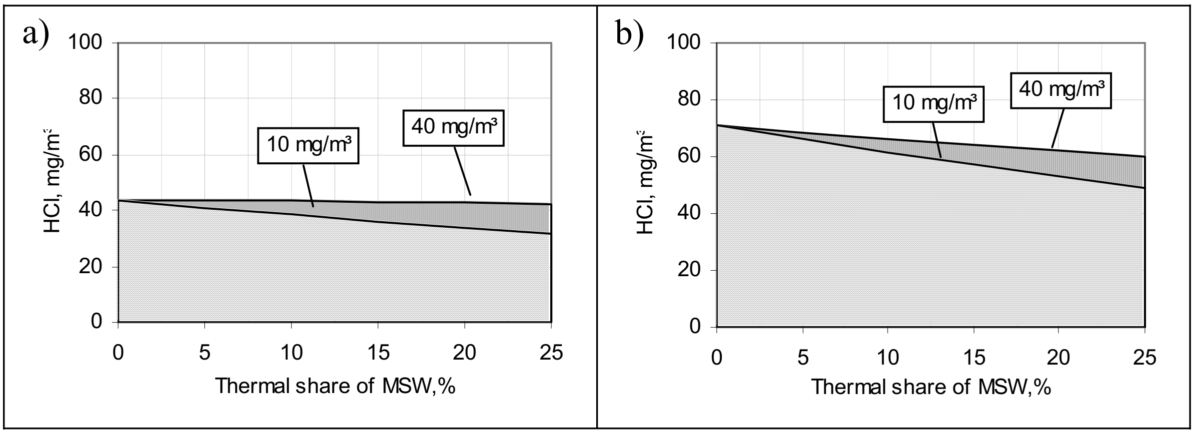


Fig. 5-8: HCl concentration, mg/m^3 a) bituminous coal B/1 b) lignite L/1

Due to the fact that the residual HCl concentration of the waste flue gases behind the partial flue gas treatment system is lower in comparison to those from 100% coal combustion, lower HCl concentrations in the mixed flue gases can be expected for the integrated process. The dilution effect will be even greater for coal with higher chlorine content.

5.2.2 Fate of chlorine during SRF co-combustion

To improve the understanding of the pollutant behaviour in the co-combustion process, the fate of chlorine was investigated in detail. Experiments were performed using the electrically heated combustion reactor BTS described in chapter 4.1. Two coals, the Colombian bituminous coal B/1 and the German lignite L/3, and two SRF qualities (SRF/04 and SRF/05) were co-combusted at approx. 5 and 10% thermal share. The properties of the investigated fuels were already discussed in chapter 3.2.1. Due to their importance the concentrations of chlorine, ash, and calcium found in the investigated fuels are summarised in the following table:

Table 5-18: Chlorine, ash and calcium content of the investigated fuels

Parameter	Unit	B/1	L/3	SRF/04	SRF/05
Chlorine	$\%_{daf}$	0.05	0.05	0.34	0.64
Ash	$\%_{dry}$	15.5	9.4	12.9	11.5
Ca	$\%_{dry}$	0.20	1.34	2.75	2.47

The chlorine concentration in both SRF materials is significantly higher compared to the coals. Due to this fact, increased chlorine concentrations can be expected, even for relatively low shares of SRF. Comparing both SRF with the investigated coals it is seen that the calcium content is significantly higher. Both ash and calcium content of the SRF will influence the properties of the coal ash, and an assessment of ash bound chlorine appears necessary. In order to quantify the chlorine concentration in the combustion process, eight different test settings (or flames) were investigated. The thermal input was adjusted to approx. 6 kW for each flame. During co-combustion operation, an equivalent of approx. 5% and 10% of the thermal input into the reactor was replaced by SRF. The respective combustion settings are summarised in the following tables:

Table 5-19: Combustion settings bituminous coal B/1 – SRF

Coal, kg/h	P, kW	SRF share	SRF, kW	SRF, g/h	Ash, kg/h	P, kW
0.800	5.92	0%	0.00	0	0.12	5.92
0.768	5.69	5%	0.24	42	0.12	5.92
0.736	5.45	10%	0.47	85	0.12	5.92

Table 5-20: Combustion settings lignite L/3 – SRF

Coal, kg/h	P, kW	SRF share	SRF, kW	SRF, g/h	Ash, kg/h	P, kW
1.200	5.99	0%	0.00	0	0.09	5.99
1.152	5.75	5%	0.24	43	0.09	5.99
1.104	5.51	10%	0.48	86	0.10	5.99

The following test matrix summarises the experiments performed at the test facility, comprising of baseline tests (pure coal) and co-combustion tests, while the given percentages indicate the thermal share of SRF during each test:

Table 5-21: Test matrix

Coal	Baseline tests	SRF/04		SRF/05	
German lignite L/3	0% (no SRF)	5%	10%	5%	10%
Bituminous coal B/1	0% (no SRF)	5%	10%	-	-

During each measurement hydrogen chlorine (HCl) was measured following German VDI guideline 3480 as outlined in chapter 4.6. In order to evaluate the results process balances were calculated based on the initial fuel analysis. A detailed description of this approach can be found in Appendix B-0. Fig. 5-9 summarises the chlorine measurements for baseline and 5/10% thermal share for the bituminous coal B/1. In Fig. 5-9a the measured gas concentrations are compared with calculated maximum values. In this context, the recovery rate is the ratio of measured vs. maximum gas concentration. Fig. 5-9b shows the measured results for the ash constituents Ca, C, and Cl.

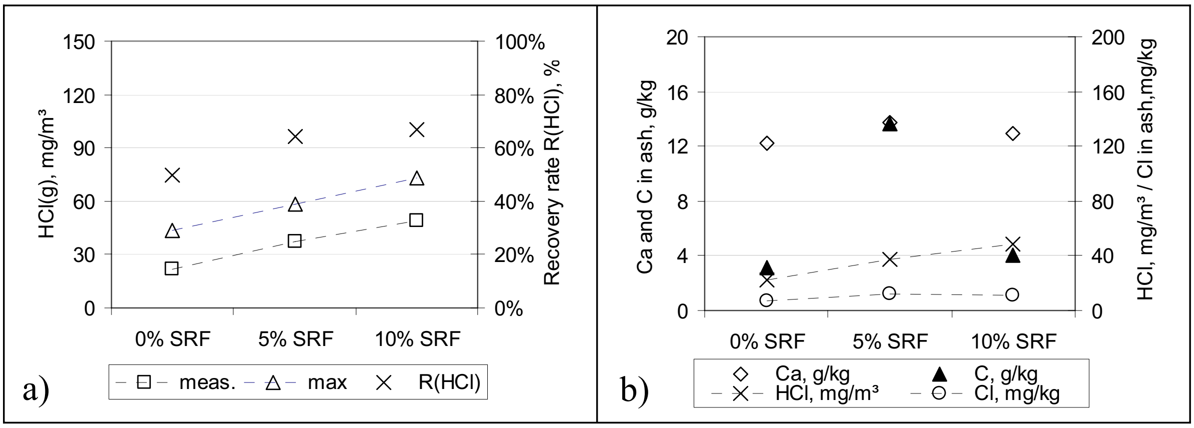


Fig. 5-9: SRF/04.B/1 a) HCl balance b) HCl/Cl vs. carbon and calcium

Fig. 5-9a indicates that the HCl concentration increases from approx. 22 mg/m³ (baseline) to 50mg/m³ (10% SRF share). Compared to the maximum concentrations the measured values are lower for all investigated test settings. The recovery rate ranges between 50 and 70%. Fig. 5-9b shows that the chlorine concentration in the fly ashes does not increase as a consequence of the increased chlorine concentration in the gas phase. The calcium content of the fly ashes is comparable for all tests. Furthermore, it can be seen that the high unburnt carbon content of the 5% flame does not change this behaviour. Fig. 5-10 summarises the results of the chlorine measurements for the German lignite L/3:

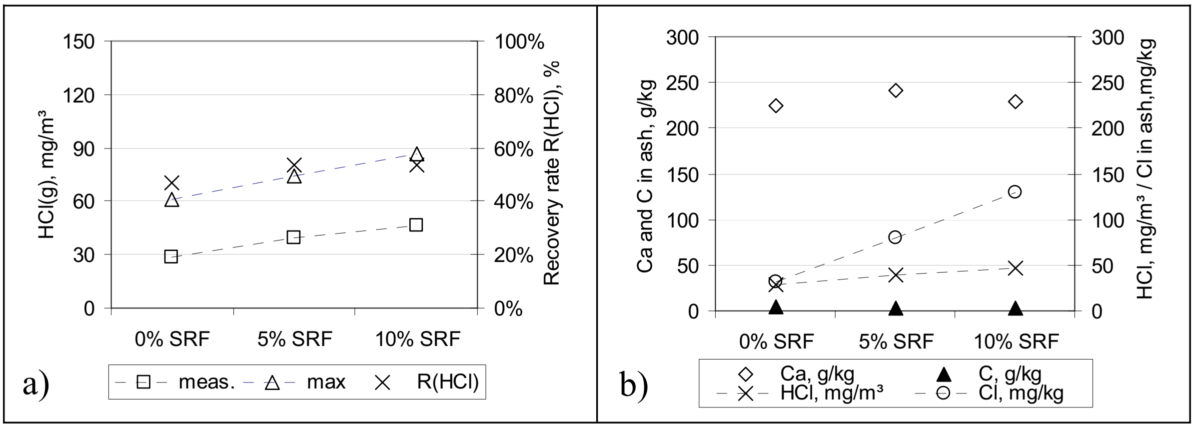


Fig. 5-10: SRF/04.L/3 a) HCl balance b) HCl/Cl vs. carbon and calcium

The measured chlorine concentration increases from approx. 30 mg/m³ (baseline) to 50 mg/m³ (10% SRF share). Comparable to the results found for the bituminous coal, the measured chlorine concentrations are significantly lower compared to the calculated maximum values. The recovery rates range between 47 and 54%. Contrary to the experiments with the bituminous coal, the chlorine concentration in the fly ashes is significantly increased. The calcium content of the fly ashes does not change significantly, and unburnt carbon is almost zero. Fig. 5-11 shows the results for the system SRF/05 – L/3:

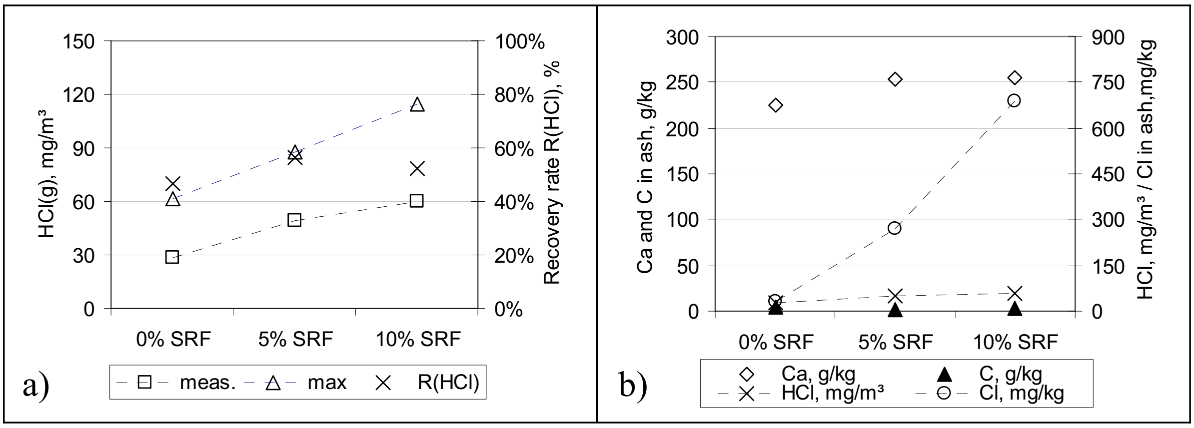
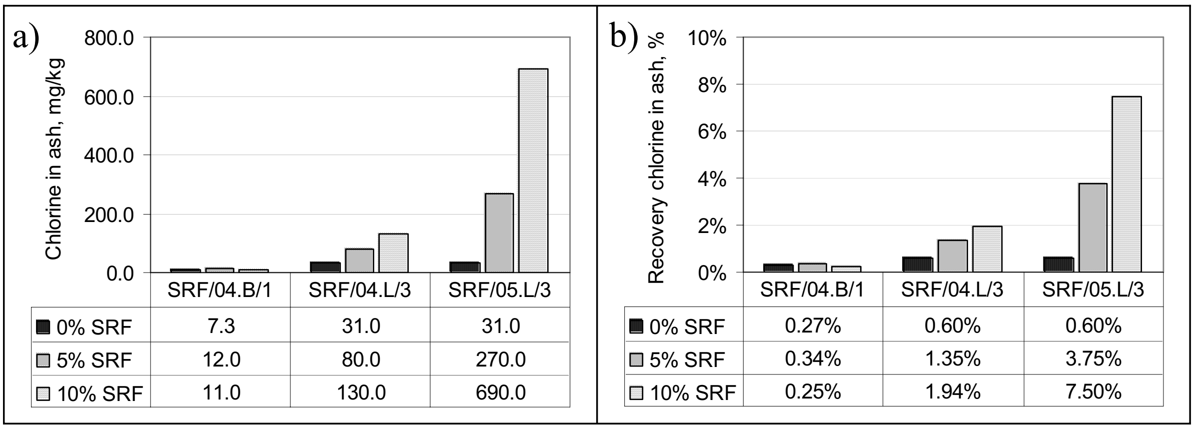


Fig. 5-11: SRF/05.L/3 a) HCl balance b) HCl/Cl vs. carbon and calcium

The measured chlorine concentration in the flue gases is slightly higher compared to the system SRF/04 – L/3 (approx. 60 mg/m³ for 10% SRF share). This can be explained with the higher chlorine content of the SRF/05. A significant difference can be determined considering ash bound chlorine. The concentration is almost five times higher. Contrary to the results discussed before, a slight increase was determined for the calcium concentration in the fly ashes. The content of residual carbon is comparatively low. In order to investigate this behaviour more in detail, recovery rates of chlorine in ash can be calculated. The results are summarised in the following figure, where Fig. 5-12a shows the measured values and Fig. 5-12b the calculated recovery rates:



a)

	SRF/04.B/1	SRF/04.L/3	SRF/05.L/3
■ 0% SRF	7.3	31.0	31.0
▨ 5% SRF	12.0	80.0	270.0
□ 10% SRF	11.0	130.0	690.0

b)

	SRF/04.B/1	SRF/04.L/3	SRF/05.L/3
■ 0% SRF	0.27%	0.60%	0.60%
▨ 5% SRF	0.34%	1.35%	3.75%
□ 10% SRF	0.25%	1.94%	7.50%

Fig. 5-12: a) Chloride in ash, mg/kg b) Recovery of chlorine in ash, %

For the bituminous coal the amount of chlorine in the fly ash is almost constant and ranges between 0.25 to 0.35%, obviously not influenced by an increased chlorine concentration in the flue gas. This finding can be explained with the low calcium content of the hard coal, which is approx. 1/6 that of the lignite.

Therefore the situation found for the lignite is completely different. The ashes apparently have a certain capability to adsorb a significant amount of the additional chlorine, although this absorption capacity does not explain the significant increase of ash bound chlorine determined for the experiments with SRF/05.

5.2.3 Comparison and evaluation of results

Considering the co-combustion process, the expected increase of gaseous chlorine as a result of higher chlorine content in the SRF materials could be verified, although the HCl concentration alone allows no assessment of a potential increased corrosion risk. The recovery rates for the chlorine in the gas phase - measured as hydrogen chloride - ranged between 50 and 70% for all experiments performed. The reason for this is related to the ash mass balance which cannot be closed due to the unavoidable loss of ash in the ducts of the test facility. Despite of this aspect, it is generally assumed that the test results are comparable as the relevant boundary conditions (reactor temperature, power input, combustion air, etc.) were similar for each flame.

Considering the amount of chlorine bound to the ashes significant differences could be determined between the investigated hard coal and the lignite. No influence of an increased chlorine concentration in the gas phase could be determined for the bituminous coal, whereas significant increase was observed for the lignite, especially for the trials with SRF/05 (up to 8% of the initial chlorine freight). This increase cannot be explained with the chlorine concentration in the flue gas alone. It is assumed that the ashes are somehow activated, thus enhancing their capability to absorb a larger amount of chlorine. Contrary to the measured HCl in the gas phase, the increased chlorine content in the lignite ashes may be taken as an indicator for an increased corrosion risk, but a final statement towards corrosion based on the trials performed is not possible.

For the UPSWING process it can be stated that the partial flue gas treatment concept consequently limits the introduction of waste-borne chlorine into the power plant process. The residual (gaseous) chlorine concentration in the waste flue gases are in most cases lower in comparison to those of 100% coal combustion. Consequently, a dilution of the mixed flue gases is expected. Furthermore, no waste-borne ashes are introduced into the power plant process as a result of almost complete ash (and dust) removal prior to flue gas injection. Contrary to SRF co-combustion, no unwanted introduction of critical ash constituents like sodium, potassium, etc. takes place. Consequently, the risk of increased boiler corrosion is significantly lower for the UPSWING concept.

5.3 Destruction of PCDD/F

PCDD/F summarises the family of chlorinated organic compounds consisting of polychlorinated dibenzo furans (PCDF) and polychlorinated dibenzo dioxins (PCDD). PCDD/Fs build up or bioaccumulate in living tissue over time, so even small exposures may accumulate to dangerous levels. Excessive exposure to dioxin may cause a severe form of persistent acne (chloracne), which is the only known direct result of dioxin exposure at levels below the lethal dose. Other possible effects may be developmental abnormalities, birth defects, or damage to the immune system.

The basic structure of PCDD/Fs comprises of two benzene rings joined by either a single (furan) or a double oxygen bridge (dioxin). Chlorine atoms are attached to the basic structure at 8 different places on the molecule, numbered from 1 to 10 (five C-atom per benzene). There are 210 different PCDD/F congeners (75 PCDDs and 135 PCDFs). The toxicity of PCDD/F is dependent on the number and position of the chlorine atoms. Only congeners that have chlorines in the 2, 3, 7, and 8 positions have any observable toxicity. Out of the 210 PCDD/F compounds in total, only 17 congeners (7 PCDDs and 10 PCDFs) have chlorine atoms in the relevant positions to be considered toxic by the NATO/CCMS international toxic equivalent (I-TEQ) scheme [21][29].

The formation and destruction of PCDD/F in thermal processes is still of high concern, especially in case of waste incineration processes. PCDD/F are brought into the process with the waste, while average pollutant concentrations of approx. 50 to 100 ng(I-TEQ)/kg can be found [34]. Recent investigations clearly indicated that PCDD/F introduced with the waste are destroyed to a large extent as the result of an optimised combustion process. Despite of this, a new formation of PCCD/F during the process is generally possible. In a temperature range between 450 and 250°C, PCDD/Fs can be formed by a heterogeneous oxy-chlorination reaction, the de-novo synthesis [35]. The ingredients and conditions necessary to form dioxin include products of incomplete combustion (PIC), halogenides (mainly Cl^-, but also Br^-), oxidising atmosphere, dust load, and a catalyst, whereas copper (Cu) salts being the most effective ones. Considering waste incineration, the above-mentioned prerequisites are given. It was found that good combustion control for burnout optimization is the best and cheapest measure to minimize PCDD/F formation down to raw gas levels of 1-5 ng(I-TEQ)/m^3 [36]. This can be achieved by favourable process conditions in the burnout zone above the grate. The demands for a good burnout of the gas phase species include a minimum residence time of 2 s at temperatures above 850°C.

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[34] B. JOHNKE: "Dioxinemissionen aus Abfallverbrennungsanlagen", Ensorga-Magazin 4/5, S. 87/188, (1994)

[35] H. VOGG, L. STIEGLITZ: "Thermal Behaviour of PCDD/PCDF in Fly Ash from Municipal Incinerators", Chemosphere, 15, 1373, (1986)

[36] H. VOGG, H. HUNSINGER, A. MERZ, L. STIEGLITZ, J. VEHLOW: "Head-end-Techniken zur Dioxinminderung". VDI Berichte 895, 193, (1991)

The oxygen content has to be at least 6 vol.-%, and favourable mixing conditions have to be ensured.

The situation in terms of PCDD/F in a modern waste incinerator is schematically depicted in Fig. 5-13. The PCDD/F raw gas level downstream of the boiler can be kept below 5 ng(I-TEQ)/m³, which allows an easy compliance with the meanwhile almost worldwide found emission standard of 0.1 ng(I-TEQ)/m³. This low emission number has reduced the contribution of waste incineration to the total annual dioxin emission in Germany from 400 g(I-TEQ) in 1990 to less than 0.4 g(I-TEQ) in 2000 which is < 1% [37].

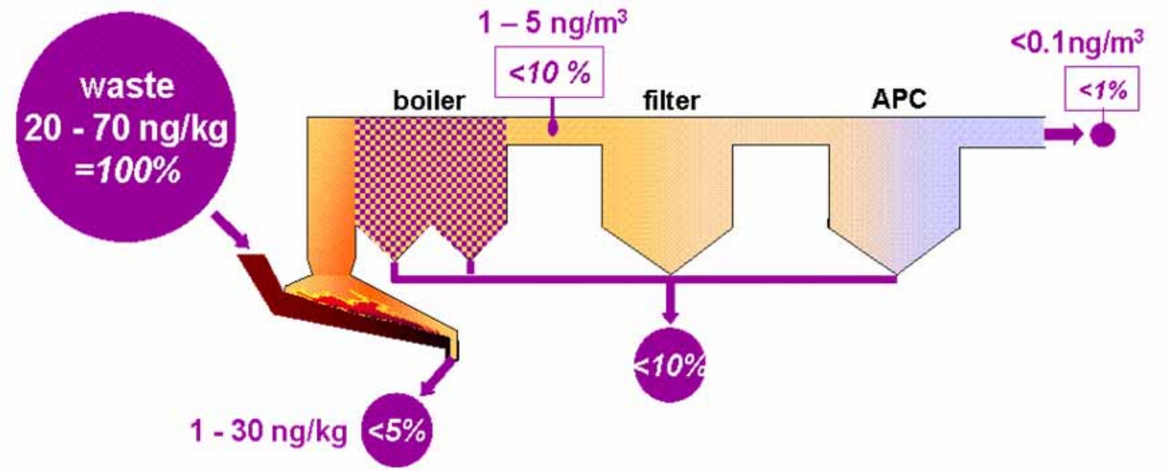

Fig. 5-13: PCDD/F mass balance in a state-of-the-art waste incineration plant [38]

The main solid residue stream, bottom ash, carries low PCDD/F loads [38] which are not far above the typical concentrations found in Central European soil [39]. Filter ashes and other gas cleaning residues are classified as toxic waste and require special disposal or inertisation measures.

Considering pulverised coal combustion, dioxins are particularly no major problem, where only very low PCDD/F concentrations (< 0.01 ng(I-TEQ)/m³) have been found [40]. In comparison to waste incineration, the inventory of chlorine in coal is much lower and that of sulphur much higher. The resulting high SO_2/HCl ratio in the flue gases reduces the efficiency of the oxy-chlorination reaction and suppresses PCDD/F formation [41].

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[37] Umweltbundesamt (UBA), (2005)

[38] J. VEHLOW: “The ash characteristics and treatment methods for the destruction of dioxin- like compounds in incinerator residues”, International workshop on the reduction and control of PCDD/F from combustion, Hangzhou, China, October 18-20, (2004)

[39] H. FIEDLER: „Dioxine in Produkten und Abfällen“, VDI Berichte 1298, 231, (1996)

[40] H. HAGENMAIER, R. BEISING: “Untersuchung von Kraftwerksrauchgasen auf polychlorierte Dibenzodioxine und Dibenzofurane“, VGB Kraftwerkstechnik, 69, 1024, (1989)

[41] R.D. GRIFFIN: “A New Theory of Dioxin Formation in Municipal Solid Waste Combustion”, Chemosphere, 15, 1987, (1986)

Flue gas integration is one crucial aspect of the UPSWING concept. The flue gases are partially cleaned, where PCDD/F and other organic micro-pollutants bound to particles are removed in the ash filter system. The residual PCDD/F in the flue gases miss active adsorption centres, leading them to stay in the gas phase and being transferred into the furnace of the connected power plant. However, the introduction of PCDD/F into the power plant furnace, regardless if as an ingredient in MSW or along with the pre-cleaned flue gas, should cause no problems, as they should not survive in a well-controlled combustion process. As a result of the high combustion temperatures, the destruction yield should almost be complete. To confirm these expectations, experiments on the stability of PCDD/F have been conducted in two test facilities at IVD, the pulverized coal combustion reactor BTS and the fluidized bed reactor ELWIRA. The trials have been conducted in close cooperation with the Institute of Technical Chemistry of Forschungszentrum Karlsruhe (Germany) within the scope of the European Research Project UPSWING [42].

In terms of SRF materials detailed information towards PCDD/F levels in the material are not available. Some information could be acquired for PCP/PCB, indicating levels several magnitudes lower (10 to 100) compared to those found in MSW. Within the scope of the Thermie programme [43], which involves the combustion of refused derived fuel (RDF; a former synonym for SRF) in large utility boilers, the secondary fuel has been investigated towards 16 different PCDD/F, although none of them could be detected in the examined samples. Assuming minimum PCDD/F concentrations in the SRF materials, a question would be if co-combustion of SRF leads to increased PCDD/F emissions, newly formed in the boiler as a result of incomplete combustion.

5.3.1 PCCD/F destruction potential of the UPSWING process

Base fuel used during the experiments was the Polish lignite L/1. The coal was spiked with the non-toxic isomer 1, 2, 3, 4, 7 - pentachlorodibenzo-p-dioxin (P5CDD). In addition, the thermal more stable hexachlorobenzene (HCB) was adsorbed on the coal in order to verify the results for PCDD/F. The concentrations of the test materials are summarised in the following table:

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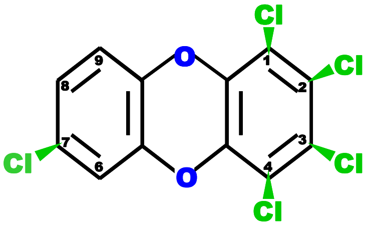
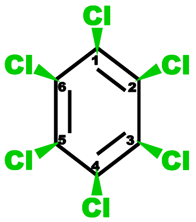
[42] J. VEHLOW, Th. HILBER, H. HUNSINGER, K. JAY, S. KREISZ, J. MAIER, H. SEIFERT: „Dioxinzerstörung im UPSWING-Prozess“, In: Optimierung der Abfallverbrennung 2 (Hrsg.: K.J. Thomé-Kozmiensky, M. Beckmann), TK-Verlag, Neuruppin, 651-662, (2004)

[43] THERMIE-PROGRAMME: „Combined combustion of RDF and coal in large utility boilers“, Project SF00115/96/IT/AT/DK, Final report, (2002)

Table 5-22: Test materials and concentrations in the spiked coal [42]

Compound	1,2,3,4,7 pentachloro dibenzo-p-dioxin	hexachlorobenzene (HCB)
Molecular structure		
Concentration in the spiked coal	1200 ng/g ± 20%	120000 ng/g ± 20%
Maximum concentration in the raw gas	9000 ng/m³	900000 ng/m³

The experiments have been conducted with a coal feed of 1 kg/h. Sampling was conducted for at least 1.5 hours per test run. The concentration of spiked coal in the fuel was adjusted to establish a theoretical raw gas concentration of 9000 ng/m³ of P5CDD and of 900000 ng/m³ of HCB. The reactor temperature of the BTS reactor was set to 1100°C, the fluidised bed reactor was operated at 850°C. Under the given process conditions, the flue gas residence time in the BTS is 3 to 4s. During the tests artificial flue gas was injected at ratios between 10 and 20%, calculated as the share of MSWI flue gas to the flue gas flow rate generated by the coal. The fluidized bed reactor was operated in bubbling and circulating mode with flue gas residence times of 5.5 and 3.5s respectively. A total of six flames (or test settings) were investigated. Reference tests were conducted for all operational modes (see table Table 5-23):

Table 5-23: Test program performed

Code	Test facility	Flue gas ratio
BTS-1, BTS-2, BTS-3	BTS	0% (reference)
BTS-FG-10	BTS	10%
BTS-FG-20	BTS	20%
FB-B	ELWIRA, bubbling	0% (reference)
FB-C	ELWIRA, circulating	0% (reference)
FB-C-FG-20	ELWIRA, circulating	20%

At the BTS facility, fly ash was sampled in the heated candle filter system, while (gaseous) PCDD/F was measured behind the filter. At the ELWIRA facility, fly ash was sampled in the cyclone and the ceramic candle filter. Comparable to the BTS trials, (gaseous) PCDD/F was measured behind both filters. When PCDD/Fs are fed into the combustion chamber, the most likely reaction in the hot and oxidizing environment is the destruction of their molecular structure. The main product is supposed to be CO_2. Another potential reaction type would be a de-chlorination or a chlorine exchange reaction. Furthermore, the de-novo synthesis has to be taken into account as the flue gases are slowly cooled down in the flue gas path of both reactors. A new formation of PCDD/F might result in the appearance of all PCDD/F homologues in the raw gas. These three options make it difficult to calculate a decomposition yield for the added P5CDD. Therefore it was decided to analyse all samples for tetra- to octa-homologues of PCDD and PCDF as well as for di- to hexachlorinated benzenes and the di- to pentaphenols.

Experimental results

The measured gas concentrations of PCDD/F are shown in Fig. 5-14. The diagrams show the presence of all homologues, although none of them is dominating, especially not the pentachlorinated ones, which were brought into the process via the spiked coals. Both, the results for PCDD and PCDF show a comparable scattering. The measured concentrations are close to the determination limit; therefore the total error is expected in the range of 20- 50%.

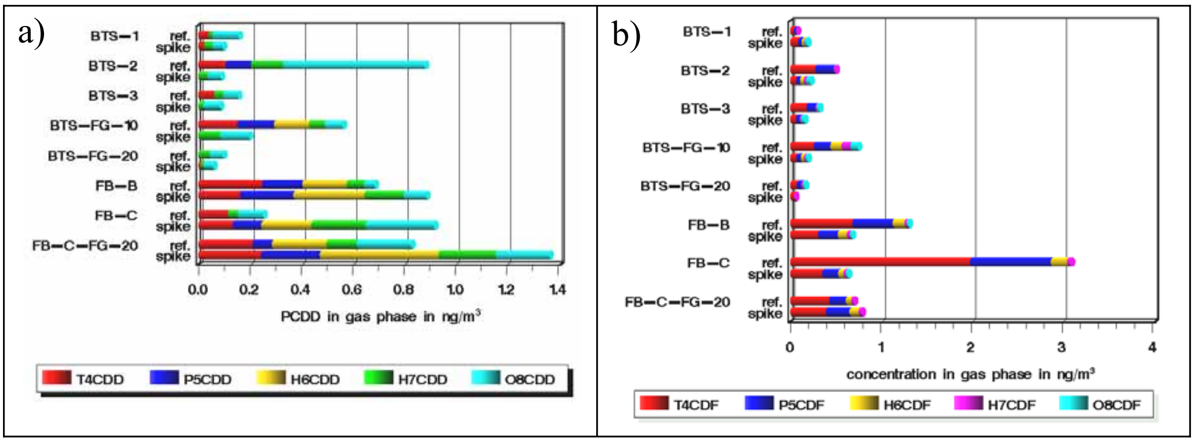


Fig. 5-14: Homologues in the gas phase a) PCDD b) PCDF

The results indicate no significant differences between the reference tests and those with spiked fuel. A slight tendency of higher PCDD concentration might be seen for the trials at the fluidised bed reactor. Fig. 5-15 shows the distribution of PCDD/F homologues in the filter ashes of both test facilities. Comparable to the results for gaseous PCDD/F, the concentrations found in the ashes are extremely low and close to the detection limit.

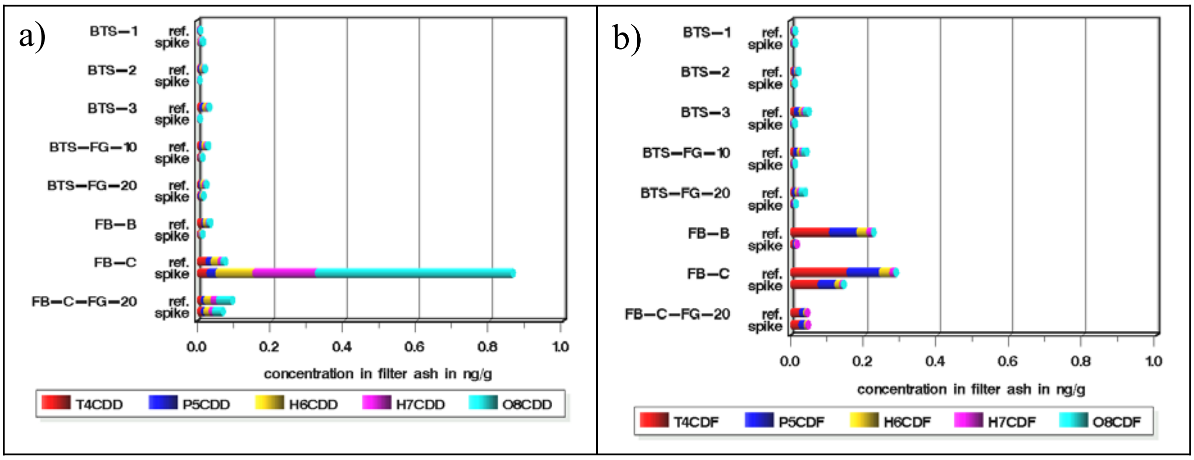


Fig. 5-15: Homologues in the filter ashes a) PCDD b) PCDF

The high PCDD concentration of the FB-C spike test is considered as an outlier since comparable concentrations have not been found in the other spike tests. It is therefore assumed that this result is caused by an analytical error. The knowledge of the gas and ash mass flows allows the calculation of the total inventory of PCDD/F in the raw gas. Fig. 5-16 shows sum concentrations of PCDD and PCDF in the raw gas of the test facilities:

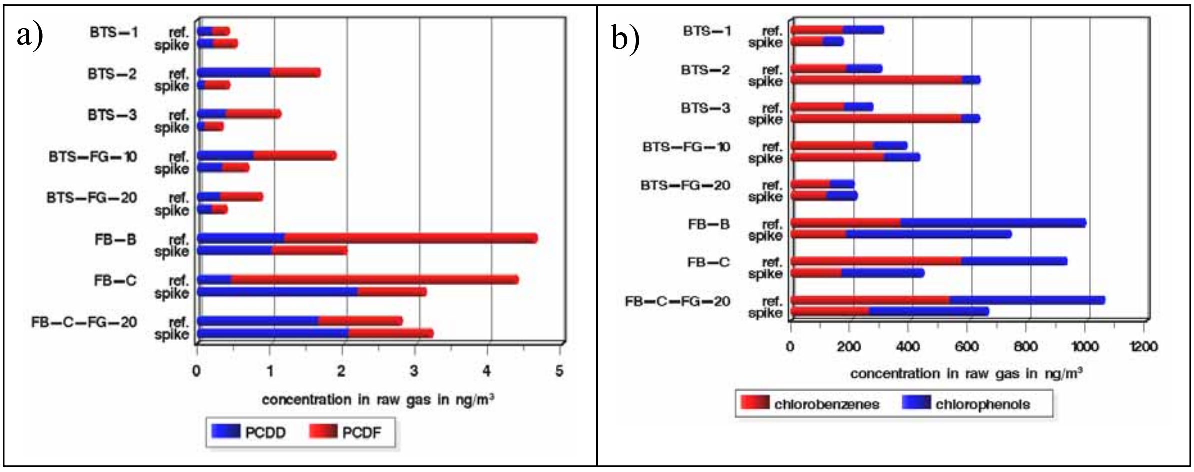


Fig. 5-16: Sum concentrations of (gaseous) a) PCDD and PCDF b) Chlorobenzenes and -phenoles

As already found for the concentrations of PCDD and PCDF in the single compartments (homologues), there is also no distinct difference recognized in the summarized raw gas concentrations of all tests. The maximum total concentration was 4.7 ng/m³ and that result was found in a reference test. If - as a conservative estimate - it would be considered that all PCDD/F were decomposition products of the 1,2,3,4,7 pentachlorodibenzo-p-dioxin spike, a minimum destruction efficiency of approx. 99.9 % could be calculated. In fact the tests documented that PCDD/Fs are totally destroyed in a well-controlled combustion process. The simultaneous appearance of all homologues of PCDD and PCDF, especially the appearance of homologues with more chlorine atoms per molecule than the starting compound, points out that these compounds are most likely formed in the flue gas path of the test facilities. The responsible formation reaction should be the de-novo-synthesis since all ingredients are present, although on a very low level. Considering HCB, there is the typical scattering of results with no indication of a significant influence of the spike. In the BTS tests more chlorobenzenes than chlorophenols are formed whereas in the ELWIRA tests the chlorophenols are slightly prevailing. All analysed concentrations sum up to slightly more than 0.1% of the spiked HCB. Hence, as for the PCDD/F also for HCB it is a fair statement that a well-controlled combustion process guarantees a thermal destruction potential which can from a technical point be called complete.

5.3.2 PCDD/F destruction potential during SRF co-combustion

It was mentioned beforehand that only insignificant PCDD/F concentration are expected in SRF. As a consequence of the high combustion temperatures in the furnace a possible PCDD/F input should be destroyed. Nevertheless, a possible source for additional emissions might be the de-novo-synthesis as a result of incomplete combustion. To clarify this question, the results acquired during a large-scale co-combustion campaign at a lignite-fired power plant should be briefly discussed. During these trials, stack emissions have been measured according to German 17th BImSchV [44], which is the national German directive in accordance with European Directive 2000/76/EG on waste incineration. The thermal share of SRF combusted during the trials was 2 respectively 4%. The relevant results are summarised in the following table:

Table 5-24: Results from large-scale co-combustion trials

	Baseline	Co-combustion	Legal limit
Dust, mg/m³	3,0	1,5	30
Total carbon, mg/m³	0.3	0.5	20
Unburnt carbon, %	0.8	0.5 - 1.0	-
PCDD/F, ng/m³	< 0.001	< 0.001	0.1

The table shows the results of baseline coal and co-combustion operation of the flue gas stack emissions for dust, total carbon, and PCDD/F. Furthermore, it shows the percentage of unburnt carbon in the fly ash. Additionaly, the limits according to 17th BImSchV are included. First of all, it can be stated that the resulting PCDD/F emissions remained well below 0.001 ng(I-TEQ)/m³ during baseline and co-combustion operation, which is two magnitudes below the legal limit of 0.1 ng(I-TEQ)/m³. Considering the other parameters, it can be seen that the levels stay in the range of standard operation. The results found for PCDD/F are in line with the usual emissions found during pulverised coal combustion as discussed in chapter 5.3.

5.3.3 Comparison and evaluation of results

The destruction potential of PCDD/F in coal-fired power plants was investigated in detail for the UPSWING process. All outlet streams have been sampled and analysed for PCDD, PCDF, PCP, and PCB. In all compartments rather low concentrations of all homologues of all compound classes have been detected. Aside from the fact that in the flue gases of the ELWIRA furnace slightly higher concentrations of all compounds have been found there was no difference in the results obtained during the reference tests - those without spikes - and those when the spiked coal was burnt. The combustion experiments with spiked coal documented a destruction yield larger than 99.9% for organic compounds. Based on these results, it can be concluded that the injection of PCDD/F containing flue gases from waste incineration into a power plant causes no environmental risk. Considering SRF co-combustion, the results acquired during large-scale co-combustion trials at a lignite-fired coal boiler indicated no increase of PCDD/F emissions.

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[44] Siebzehnte Verordnung zur Durchführung des Bundes-Immissionsschutzgesetzes, Verordnung über Verbrennungsanlagen für Abfälle oder ähnliche brennbare Stoffe, 17. BImSchV, BGBl. I, S. 2545-2832, (1990), BGBl. I, S. 1950, (2001), BGBl. I, S. 1614-1633, (2003)

5.4 Trace element behaviour

Fossil fuels contain heavy metals by nature in different concentrations. They are bound to the inorganic as well as to the organic matter and represent the major part of trace substances found. Table 5-25 summarises the typical concentration of selected elements found in municipal solid waste, bituminous coal and lignite:

Table 5-25: Mean concentrations of selected elements in MSW, hard coal and lignite in mg/kg [45]

	Municipal solid waste		Bituminous coal		Lignite	
	Mean	Range	Mean	Range	Mean	Range
S	4000	1000 - 6000	7000	4000 - 15000	4000	2000 - 32000
Cl	7000	3000 - 9000	1000	100 - 3000	300	400 - 1000
As	7	2 - 20	20	1 - 60	n.a.	n.a.
Cd	10	3 - 20	1	< 0.1 - 2.5	n.a.	n.a.
Hg	3	0.5 - 10	0.12	0.1 - 0.6	0.2	0.01 - 1
Pb	700	400 - 1500	20	10 - 60	n.a.	n.a.

It is obvious that MSW contains significantly higher amounts of heavy metals in comparison to coal, especially toxic elements such as cadmium (Cd), mercury (Hg), or lead (Pb). Released during the combustion process, abating direct emissions of such environmental critical substances is necessary. This could be achieved by a good control of the process and by suitable flue gas cleaning technologies as outlined in chapter 2.1.1.

Depending on the design and operation conditions of the combustion process (fuel input, firing system, and flue gas cleaning system), heavy metals are distributed in different power plant products/residues like fly ash, bottom ash, slag, FGD products and flue gas. In the power plant process these are mainly bottom ash, fly ash and the flue gas. Enrichment in FGD product gypsum is also possible, although it was determined in several investigations that heavy metals concentrate in the fly ashes and the amount found in gypsum and flue gas is usually below 3% of the initial freight [46]. Table 5-26 summarises usual concentrations of heavy metals in fly ashes from different technical applications.

Comparing trace elements found in MSWI fly ashes with conventional power plant ashes, the concentrations are likely to be 10 to 100 times higher. In this context it has to be emphasised that the partial flue gas cleaning concept of the UPSWING process basically omits the introduction of fly ash into the power plant process as outlined in chapter 3.1.1. Contrary to this, co-combustion of SRF may lead to an enrichment of (certain) elements in the power plant process. Accordingly, fly ash has been selected to study the effects of co-combustion on

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[45] J. VEHLOW, H. HUNSINGER, S. KREISZ, H. SEIFERT: „Das UPSWING Verfahren - Der Schlüssel zur kostengünstigen Abfallverbrennung“, Forschungszentrum Karlsruhe, Nachrichten 32, S. 201ff, (2000)

[46] Ch. MARTEL: „Brennstoff- und lastspezifische Untersuchungen zum Verhalten von Schwermetallen in Kohlenstaubfeuerung“, PhD – thesis, IVD, University Stuttgart, (2000)

trace element behaviour, although not all elements could be considered. Therefore, it was decided to focus on some characteristic elements with different physical and chemical behaviour, especially in terms of their volatility, but also in terms of their toxicological importance.

Table 5-26: Heavy metal concentrations in different fly ashes

Origin →	MSWI flue gases, raw [47]	MSWI fly ashes [48]	Hard coal fly ashes [49]	Fly ashes lignite [50]
Element ↓	mg/m^3_N	Ppm	ppm	ppm
Cd	0.3 - 2.5	100 - 1400	0.2 - 7	0.1 - 0.45
Pb	10 - 60	2500 - 25000	23 - 817	3 - 100
Cu	10 - 50	50 - 5000	38 - 613	2 - 20
Zn	10 - 150	5000 - 100000	47 - 1483	11 - 42
Hg	0.4 - 0.7	1 – 10	0.5 - 0.7	0.02 - 0.64
Ni	0.2 - 1.5	100 - 1000	46 - 301	11 – 51

Detailed investigations towards the volatility of trace elements were done by Clarke [51] and Berger [52] for pulverised coal firings. It was found that the elements can be classified according to their volatility. The properties of the elements in the different groups can be summarised as follows:

- Group 1 elements represent low-volatile elements. Evaporation processes are negligible; therefore the amounts found in the combustion residues are usually similar to those in the coal.
- Group 2 elements represent elements which usually evaporate and condensate on fly ash particles. Within this group an enrichment of the heavy metal concentration in the fly ash can be expected.
- Group 3 are high-volatile elements. Due to the low boiling points, the elements are completely evaporated into the gas phase with a low tendency for condensation. The concentration of these elements in the combustion residues is usually lower compared to the initial concentrations found in the fuel.

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[47] Rat von Sachverständigen für Umweltfragen: Sondergutachten Abfallwirtschaft, Lehrstuhl für Feststoff- und Grenzflächenverfahrenstechnik, Metzler-Poeschel, Stuttgart, (1991)

[48] L. BIRNBAUM, U. RICHERS, W. KÖPPEL: „Untersuchung der physikalisch / chemischen Eigenschaften von Filterstäuben aus Müllverbrennungsanlagen (MVA)“, Forschungszentrum Karlsruhe, Technik und Umwelt, Wissenschaftliche Berichte FZKA 5693, (1996)

[49] B. ZESCHMAR-LAHL: „Stoffflussanalyse als Planungsinstrument für den Einsatz von Ersatzbrennstoffen“, Oyten, (2004)

[50] RWE Aktiengesellschaft: „Handbuch der Verwertung von Braunkohlefilteraschen in Deutschland“, (1995)

[51] L.B. CLARKE, L.L. SLOSS: ”Trace elements - Emission from coal combustion and gasification”, IEA Coal Research”, IEACR/49, London, (1992)

[52] R. BERGER, H.J. KRABBE: „Simulation der Schwermetallemissionen eines Steinkohlekraftwerks“, Dortmund, VGB Kraftwerkstechnik 9/98, (1998)

The following figure shows the classification system for trace metals as described above:

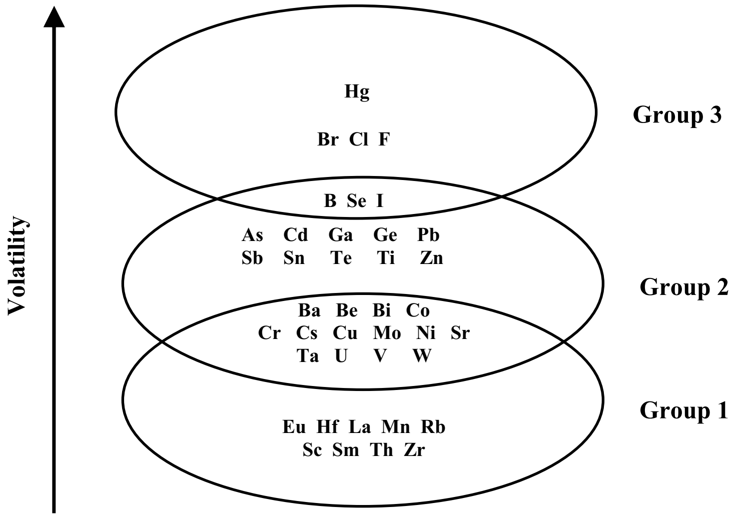

Fig. 5-17: Classification of heavy metals according to their volatility [51]

Based on this classification system, the following elements were selected as representatives for the individual groups: Mercury (Hg), Cadmium (Cd), Lead (Pb), Copper (Cu), Nickel (Ni), and Zinc (Zn). Among the selected elements, the toxic and volatile element mercury is of special relevance. According to the European position paper on mercury, approx. 340 tonnes are emitted annually from anthropogenic sources in Europe [53]. Generally, mercury emissions from coal-fired boilers can be classified into three main forms [54]:

- Particle-associated mercury, Hg(p),
- Gaseous divalent mercury, $Hg^{2+}X(g)$, $X = Cl_2(g)$, $SO_4(s)$, O(s,g) and
- Gaseous elemental mercury, $Hg^0(g)$.

During the combustion process mercury is completely transferred to the gas phase. Above 800°C only elemental mercury $Hg^0(g)$ is present. Below 500°C gaseous mercuric chloride - $HgCl_2(g)$ - is generally considered to be the dominant $Hg^{2+}X(g)$ form, but the formation of $HgCl_2$ is not only temperature dependent. The residence time, chloride concentration, but also other flue gas components and ash constituents have a major influence on the formation of $HgCl_2(g)$. Gaseous $HgCl_2(g)$ is highly water soluble and removal in the FGD system is likely, like shown in [55] and [56]. Especially in power plants

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[53] European Position Paper on Hg, http://europa.eu.int/comm/environment/air/ background.htm

[54] C. ZYGARLICKE, K. GALBREATH: "Flue gas interactions of mercury, chlorine, and ash during coal combustion", Energy & Environmental Research Center, University of North Dakota, (2001)

[55] H. THORWARTH: "The EU-Project TOMERED – Results of Trace Metal Investigations", Proceedings of the IFRF Topic-Oriented Technical Meeting 28, Salt Lake City, USA, (2006)

[56] J. TEMBRINK, H. THORWARTH, A. DIECKMANN: "Mercury Behaviour in Coal-Fired Power Plants with Sewage Sludge Co-Combustion", Proceedings of the 3rd International Experts´ Workshop – Mercury Emissions from Coal, Katowicze, Poland, (2006)

equipped with a FGD system, an increase of ionic mercury would therefore be beneficial. Laudal [57] for example determined during comparable investigations that in presence of standard flue gas components like fly ash, SO_2, HCl, NO/NO_2 and Cl_2 up to 50% of the total mercury is transferred to $HgCl_2(g)$. Therefore, the investigation of mercury behaviour in a real combustion environment appears to be favourable.

Taking the above-made findings into account, it is obvious that chlorine should have a significant influence on mercury speciation. Considering the UPSWING process, the partially cleaned waste flue gases contain gaseous chlorine, although in low concentrations. According to the findings in chapter 5.2, a dilution of the coal flue gases can be expected. Contrary to this, it was found that direct SRF co-combustion increases the availability of chlorine in the flue gases. Therefore, an assessment of this potential influence on mercury and towards mercury speciation has to be investigated.

5.4.1 Influence of waste flue gas integration on trace element behaviour

One of the major benefits of the UPSWING concept is the almost complete removal of trace elements in the ash filter system prior to flue gas injection. The resulting element concentration in the waste flue gases is therefore expected to be considerably low. Table 5-27 summarises investigations towards the removal capacities of modern fly-ash removal systems such as electrostatic precipitators:

Table 5-27: Removal capacity of ESP

Element	Removal capacity of ESP		
	Easom [58]	Veaux [59]	Martel [46]
As	81.0 %	98.9 %	97.3 %
Ba	99.8 %	n.a.	n.a.
Cd	99.2 %	98.5 %	97.3 %
Cr	99.2 %	n.a.	n.a.
Co	99.3 %	n.a.	n.a.
Cu	99.6 %	n.a.	n.a.
Ni	98.2 %	98.2 %	95.0 %
Pb	n.a.	99.0 %	97.2 %
Zn	n.a.	98.3 %	n.a.
Hg	<20 %	n.a.	74.2 %

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[57] D.L. LAUDAL, T. BROWN, B. NOTT: "Effects of flue gas constituents on mercury speciation", Grand Forks, USA, (1998)

[58] B.H. EASOM, L.A. SMOLENSKY, S.R. WYSK: "Electrocore separator for particulate air emission", Clearwater, USA, (1998)

[59] CH. VEAUX, U. KARL, O. RENTZ: „Systematische Ermittlung von Schwermetallströmen in Kraftwerken", DFIU, Karlsruhe

With the exception of mercury and possibly arsenic as determined by Easom, the removal efficiency is generally high, in most cases larger than 99%. Despite this high removal efficiency, the influence of flue gas injection on trace elements was investigated in accordance with the planned, technical evaluation of the UPSWING process. In this context the influence of an additional water input was identified as highly relevant. The acid scrubbing stage leads to a significant increase of water in the flue gases as a result of operating temperatures between 60-70°C. Full saturation can be expected, increasing the flue gas volume to some 30 to 40%. The additional water load reduces the maximum combustion (or flame) temperatures in the coal boiler. This might result in a reduced evaporation rate of trace elements. On the other hand, the total flue gas volume is increased, leading to shorter residence times in the flue gas ducts, which leads to reduced condensation of heavy metals on fly ash particles. Another important question is of course the fate of additional mercury in the UPSWING process, especially towards its chemical speciation. Despite all technical solutions or advanced retention technologies, it is expected that a certain fraction of the metal passes the partial flue gas treatment in form of gaseous metallic mercury.

To investigate the influence of flue gas injection, experiments were performed at the BTS combustion reactor (refer to chapter 4.1). MSWI waste flue gases were artificially generated and injected into the main combustion zone via the burner air. The following parameters were investigated in detail:

- **Chlorine concentration**: The influence of an additional hydrogen chloride input on the behaviour and speciation of mercury was investigated for different additional chlorine loads of the waste flue gas.
- **Process temperature**: The flame temperature is reduced by the injection of relatively cold and wet flue gases. To investigate these effects, the combustion reactor was operated and measurements were performed at two different wall temperatures (1100/1300°C).
- **Additional water load**: To investigate the influence of the scrubbing process, an additional flue gas water load was evaporated and injected into the combustion reactor. In the following discussion “wet” always refers to the injection of additional water - always in combination with the flue gases - whereas “dry” refers to the baseline case (100% coal combustion, no flue gas injection, no additional water).
- **Available oxygen** (influence of air staging): The influence of air staging (as a primary NO_x reduction measure, refer to chapter 5.1) on the behaviour of heavy metals was investigated reducing the available oxygen in the main combustion zone. During these investigations, technically applied air excess air levels of 0.95 and 0.85 were taken into account. Burnout air was injected at a fixed position in 1.4m distance from the burner, increasing the excess air level to 1.20.

Two coals were used during these experiments: The Polish lignite L/1 and the Columbian bituminous coal B/1. Both fuels were analysed towards the selected heavy metals, as shown in the following table:

Table 5-28: Heavy metal content of the base fuels

Fuel	Unit	Hg	Cd	Zn	Ni	Cu	Pb
Bituminous B/1	mg/kg	0.12	0.26	93	12	7.7	2.9
Lignite L/1	mg/kg	0.40	< 0.15	29.8	18.5	11.4	10.6

The mercury content of the lignite is approx. 4 times, the lead content approx. 3 times the content found in the bituminous coal. Cadmium and zinc concentration in the bituminous coal is considerably higher than in the lignite. Copper and nickel concentration are more or less comparable. It was mentioned before that the ash content of the fuel is a further important parameter influencing the behaviour of trace elements in the process as the primary condensation surface. The investigated lignite has an ash content of $35.5\%_{dry}$. If the complete ash is found in the gas phase, fly ash concentration would be 58.2 g/m^3. The ash content of the hard coal, 15.5 $\%_{dry}$, is significantly lower. Consequently, the maximum ash concentration in the flue gases is lower as well (16.3 g/m^3). As a consequence, the higher ash content of the lignite offers a favourable condensation surface.

a) Mercury

Gaseous mercury was measured during the trials using a continuous mercury monitor (CMM). A technical description of the device can be found in chapter 4.5.1. A speciation towards elementary $Hg^0(g)$ and gaseous $HgCl_2(g)$ was done and is available for all investigated cases. The calculation of $Hg^0(g)$, $HgCl_2(g)$ and particle bound Hg(p) was done according to the balances described in appendix B-0. Detailed results of the individual experiments are summarised in appendix C-1.

Generally, a reduction of the elemental $Hg^0(g)$ concentration in the flue gases is considered beneficial. In the power plant process, particle bound Hg(p) can be removed by the ESP together with the fly ashes and $HgCl_2(g)$ will be removed in the FGD system. The following tables summarise the results of the investigated cases for both coals in form of a parameter study based on the results summarised in appendix C-1. Parameters taken into account were process temperature, additional water load, available oxygen, and chlorine. Table 5-29 shows the results for the Polish lignite L/1:

Table 5-29: Mercury concentration in the flue gas for the Polish lignite L/1

Lignite L/1	Process temperature ⬀	H_2O ⬀	Available O_2⬀	HCl ⬀
$Hg^0(g)$	↓	↑	→	↓
$HgCl_2(g)$	↑	↓	→	↑
Hg(p)	[ ]	↓	[ ]	↓
↓ : Decrease → : No influence ↑ : Increase [ ] : Uncertain				

An increase of the process temperature leads to a decrease of $Hg^0(g)$ and an increase of $HgCl_2(g)$. A clear effect on particle-bound mercury could not be determined. Contrary to this, the amount of elementary mercury is increased for an additional water load of the flue gases, whereas both, $HgCl_2(g)$ and Hg(p), are reduced. The results found follow the theory that the formation of $HgCl_2(g)$ is reduced if more water is available, although it is expected that the additional water affects the fly ash characteristics, which itself changes the equilibrium between mercury and chlorine. Concerning the available oxygen, no influence was expected as mercury forms no oxides above 900°C. This could be verified based on the results. As expected, a higher chlorine concentration of the flue gases leads to an increase of $HgCl_2(g)$ and a decrease of $Hg^0(g)$. Particle-bound mercury Hg(p) is reduced as well. Table 5-30 summarises the results for the Colombian bituminous coal B/1, yet no experiments towards HCl were done:

Table 5-30: Mercury concentration in the flue gas for the Colombian bituminous coal B/1

Bituminous B/1	Process temperature ⬀	H_2O ⬀	Available O_2⬀
$Hg^0(g)$	↓	↑	↑
$HgCl_2(g)$	[→]	[→]	↑
Hg(p)	↑	↓	↓
↓ : Decrease → : No influence ↑ : Increase [ ] : Uncertain			

Comparable to the findings for the lignite, an increased process temperature reduces the fraction of elemental $Hg^0(g)$, although the fraction of gaseous divalent $HgCl_2(g)$ is apparently not affected. Yet, the amount of particle bound mercury increases. Comparable results were found for an additional water load of the flue gases, leading to increased share of elemental $Hg^0(g)$ in the gas phase. Contrary to the results found for the lignite, the available oxygen seems to have an effect on mercury, leading to reduced particle-bound fraction and to increased gaseous mercury emissions (both $Hg^0(g)$ and $HgCl_2(g)$).

For both coals it could be determined that the major part of the element remains in the gas phase. Only a minor amount can be adsorbed by the fly ash. The additional water of the waste flue gases seems to have the largest effect on the behaviour of mercury in the process, increasing the fraction of elemental $Hg^0(g)$. Furthermore, the injected water will reduce the combustion temperature inside the boiler, which increases $Hg^0(g)$ as well. As a consequence, the additional water input should be limited as far as possible. A possible solution might be a semi-dry scrubbing process instead of the wet acid scrubber, although this approach is not part of the (initial) UPSWING concept and has to be investigated more closely.

b) Behaviour of the elements Cd, Zn, Ni, Cu, Pb

The following section focuses on the behaviour of selected heavy metals for process conditions applicable in the UPSWING process. The investigations focus on additional water input and increased chlorine inventory. The results of the trials are summarised in Appendix C-1. Table 5-31 summarises the results in form of a parameter study for ash-bound elements during the injection of additional water, Table 5-32 the influence of additional chlorine:

Table 5-31: Parameter study (fly ash): Influence of additional water injection

Coal / Element	Cd	Zn	Ni	Pb	Cu	Hg(p)
Bituminous B/1	→	→	↓	[↓]	↓	↓
Lignite L/1	→	→	[ ]	→	→	↓
↓ : Decrease → : Comparable ↑ : Increase [ ] : Uncertain						

Table 5-32: Parameter study (fly ash): Influence of additional chlorine injection

Coal / Element	Cd	Zn	Ni	Pb	Cu	Hg(p)
Lignite L/1	→	↓	→	↓	↑	↓
↓ : Decrease → : Comparable ↑ : Increase [ ] : Uncertain						

Cadmium is a medium-volatile group 2 element. The cadmium concentration in the investigated fuels is very low (< 0.3 mg/kg). Contrary to this, the concentrations found in the fly ashes were well above the quantification limit, illustrating the tendency of this element to enrich in the fly ash. The recovery rate ranges from 70 to 110%. Cadmium forms gaseous cadmium chloride ($CdCl_2$) above 1000°C, which is converted to cadmium sulphate below 600°C. Considering this, the influence of chlorine should be neglected, as verified for the lignite. No influence of additional water could be determined for both coals.

Zinc was investigated as another medium-volatile group 2 element. The concentration found in the hard coal is 93 mg/kg, in the lignite 29.8 mg/kg. Above 1000°C zinc forms the volatile $ZnCl_2$, thus a clear influence of an increased chlorine concentration can be expected. Considering the recovery rates, the amount found in the lignite ashes are significantly higher compared to the hard coal. No influence of additional water could be determined. Contrary to this, a clear influence of chlorine was determined, leading to lower concentration Zn in the fly ash as a result of reduced condensation capacity of gaseous $ZnCl_2$.

Nickel was investigated as it represents a transition medium- to low-volatile element. The concentration found in the hard coal is 12 mg/kg, in the lignite 18.5 mg/kg. Above 1000°C, nickel forms the gaseous NiO which is transformed to different forms of $NiSO_4$ below 1000°C. The formation of $NiCl_2$ is not favoured;

therefore no major influence is expected for an increased chlorine concentration. This could be verified based on the acquired results. No clear tendency can be given for an additional water input. Considering the recovery rates, the amounts found in the hard coal are approx. 50% higher compared to the lignite ashes.

Lead is a further group 2 element. The concentration found in the hard coal is 2.9 mg/kg, in the lignite 10.6 mg/kg. Above 1000°C, lead forms gaseous PbO and $PbCl_2$. Furthermore, the formation of $PbSO_4$ can be expected. Comparable to zinc an influence of chlorine could be determined, resulting in a lower concentration in the fly ash. No major influence of additional water could be determined, but it appears that the process temperature plays an important role, especially for the hard coal. The recovery rate for 1300°C is almost twice that of 1100°C, indicating a higher evaporation rate of the element during the combustion process. Generally, the recovery rates are significantly higher for the hard coal.

Copper is, comparable to nickel, a transition medium- to low-volatile element. The concentration found in the hard coal is 7.7 mg/kg, in the lignite 11.4 mg/kg and therefore more or less comparable. No clear description about a general behaviour of the element during combustion could be found in the literature. The results show an increased concentration as a result of additional chlorine. This would indicate that the formation of $CuCl_2$ is not favoured. No influence of additional water was determined for the lignite, whereas the concentration for the hard coal slightly decreases. The recovery rates are comparable for both coals.

To summarise the experiments performed, the results show an indifferent picture. Some elements are apparently influenced by the injection of additional water, some are not. In most cases, the element content in the fly ash remains constant. Reduced concentrations were determined for Ni, Pb, and Cu, but only for the bituminous coal. Therefore, differences between the coals have to be considered as well. Only the concentration of mercury is reduced for both investigated coals. A clearer influence was found for the injection of additional chlorine. The element concentrations of Zn, Pb, and Hg are reduced, which can be explained by a (general) lesser tendency of $MeCl_x$ to condense on fly ash particles. Both Cd and Ni appear to be unaffected. Contrary to the other elements, the concentration of Cu increases. A final explanation cannot be given, although this result indicates that $CuCl_2$ is not a favoured species. Nevertheless, limiting the chlorine input into the power plant process as far as possible appears to be necessary.

5.4.2 Influence of SRF co-combustion on trace element behaviour

Thinking about thermal utilisation of waste-derived fuels, it is necessary to discuss and assess environmental aspects. Although SRF have to be produced from non-hazardous waste streams, pollutants such as heavy metals, organic pollutants, or chlorine are present within certain limits. Generally, the concentrations of these elements are expected to be higher compared to those found in coal.

Base fuels for the experiments were the German lignite L/3 and the Colombian bituminous coal B/1. Considering the SRF, two different qualities were co-combusted at 5 respectively 10% thermal share (refer to appendix C-2). Baseline tests were performed during 100% coal combustion. The following table summarises the analytical data for the investigated fuels:

Table 5-33: Heavy metal content of the investigated fuels

Fuel	Unit	Hg	Cd	Zn	Ni	Cu	Pb
B/1	mg/kg	0.12	0.26	93	12	7.7	2.9
L/3	mg/kg	0.14	< 0.15	3.9	< 3.0	3.4	< 1.5
SRF/04	mg/kg	0.10	1.36	120	9.2	35.1	24.3
SRF/05	mg/kg	0.16	0.96	308	43.6	640	58.3

Considering mercury one can see that the concentration found in the SRF is in the same magnitude of both fossil fuels (SRF/05) or even lower (SRF/04). Based on these analyses one could assume comparable emissions. The concentrations of elements such as zinc, copper, or lead, are of several magnitudes higher compared to those of coal. Consequently, an enrichment of these elements in the (mixed) co-combustion fly ashes can be expected.

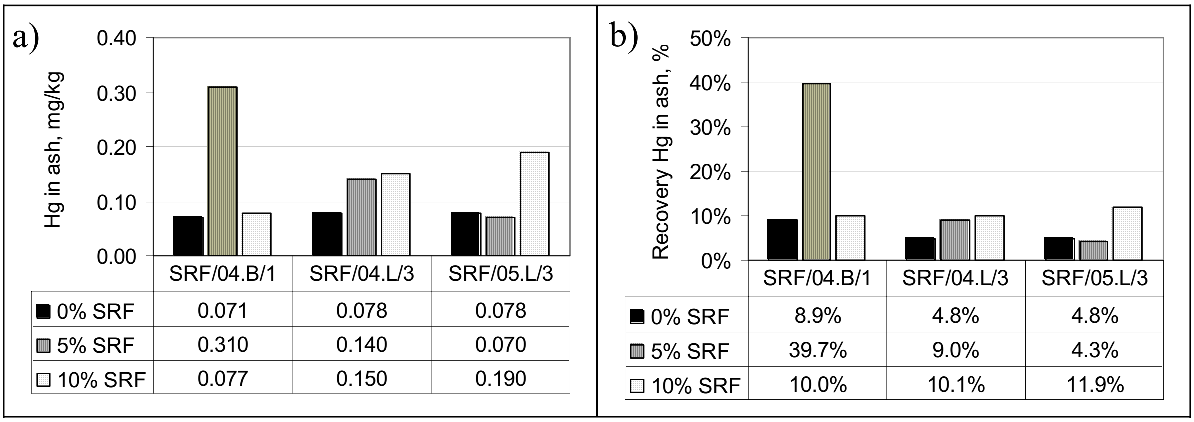
Contrary to the UPSWING concept with its complete ash removal prior to flue gas injection into the power plant process, waste-derived fuel such as SRF contains ash which is brought forward into the combustion process. This aspect has to be considered as well as it may influence both element speciation and the properties of the mixed fly ashes. In this context, an important ash constituent is calcium with a significant absorption capacity for sulphur and chlorine. Lignite ashes are mostly calcium-based, and this aspect might help to explain the analytical results. The following table summarises the ash and calcium content for the investigated fuels:

Table 5-34: Ash and calcium content of the investigated fuels

Parameter	Unit	B/1	L/3	SRF/04	SRF/05
Ash	$\%_{dry}$	15.5	9.4	18.5	13.4
Ca	$\%_{dry}$	0.20	1.34	2.75	2.47

a) Behaviour of mercury

Mercury measurements were conducted at three different positions at the BTS combustion reactor (refer to chapter 4.1), which are furnace exit (FE, 700-800°), flue gas path in front of the ash filter system (FF, 250-350°C), and behind the ash filter system (BF, 150°C). Fly ash was sampled in the candle filter system of the test facility. Contrary to the measurements performed for the UPSWING process, mercury was discontinuously measured. A description of the measurement setup can be found in chapter 4.5.2. The complete results of the trials are summarised in appendix C-2. Fig. 5-18a shows the total amount of mercury in the fly ashes, Fig. 5-18b the calculated recovery rates:



a)

	SRF/04.B/1	SRF/04.L/3	SRF/05.L/3
■ 0% SRF	0.071	0.078	0.078
▨ 5% SRF	0.310	0.140	0.070
□ 10% SRF	0.077	0.150	0.190

b)

	SRF/04.B/1	SRF/04.L/3	SRF/05.L/3
■ 0% SRF	8.9%	4.8%	4.8%
▨ 5% SRF	39.7%	9.0%	4.3%
□ 10% SRF	10.0%	10.1%	11.9%

Fig. 5-18: a) Mercury in ash, mg/kg b) Recovery of Hg in ash, %

Obviously not in line with the other results is the significant mercury content of the hard coal flame at a co-combusted share of 5%, but can be explained with the high unburnt carbon content of the fly ash (refer to Fig. 5-19a). The carbon content of the fly ashes is usually below 3 g/kg (0.3%), but during this test approx. 14 g/kg were measured. Hence, approx. 40% of the total mercury input can be found in the ash due to direct absorption of active carbon. Considering the other co-combustion ashes, a moderate increase of particle bound mercury can be determined. For the lignite the particle-bound fraction increases from approx. 5% to 10-12% for a co-combusted SRF share of 10%. In case of lignite combustion, this behaviour cannot be explained by unburnt carbon as fuel burnout is almost complete (see Fig. 5-19b):

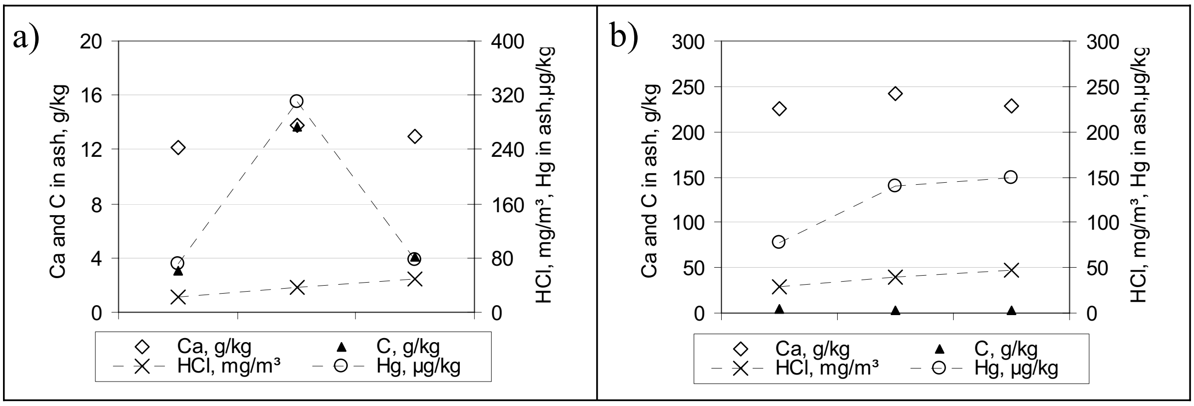


Fig. 5-19: Experimental results for a) System SRF/04.B/1 and b) System SRF/04.L/3

Considering the fact that the total mercury input is nearly constant, it is not clear which factor (or ash component) is responsible for the increasing particle-bound mercury concentration, although this effect might be connected with the higher calcium concentration of the lignite (refer to table Table 5-34).

In order to evaluate the behaviour of gaseous mercury and the influence of co-combustion on mercury speciation the system SRF/04 – Lignite L/3 will be discussed in detail. Fig. 5-20a shows the results of total gaseous mercury in µg/m³, Fig. 5-20b the calculated recovery rates of the individual measurement positions furnace exit (FE), in front of ash filter (FF), and behind filter (BF).

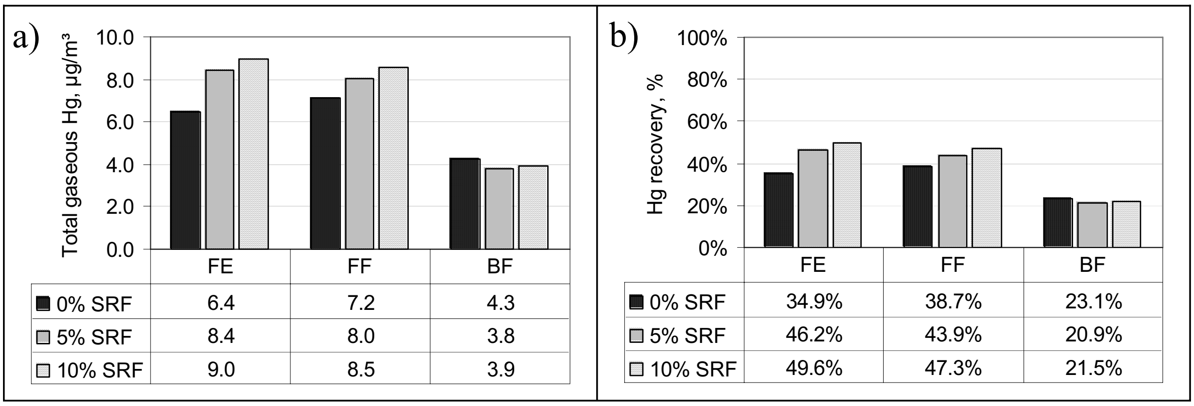


Fig. 5-20: SRF/04.IND a) Total mercury, µg/m³ b) Recovery of gaseous Hg, %

The results indicate an increasing mercury concentration in the flue gases in front of the ash filter with increasing share of SRF. Considering the initial mercury concentration in the investigated fuels this result was not expected (no or only insignificant change of total mercury was expected; refer to Table 5-33). The concentrations found behind the fly ash filter are significantly lower. It is therefore assumed that a certain mercury fraction is absorbed by the fly ash. Furthermore, the recovery rates presented in Fig. 5-20b show comparable recovery rates behind the filter, while the recovery rates at the furnace exit and in front of the filter increase during co-combustion. It is therefore assumed that the measured concentrations are only apparently higher as a result of this higher recovery rate. To summarise the results, Fig. 5-21 shows the mercury speciation along the flue gas path.

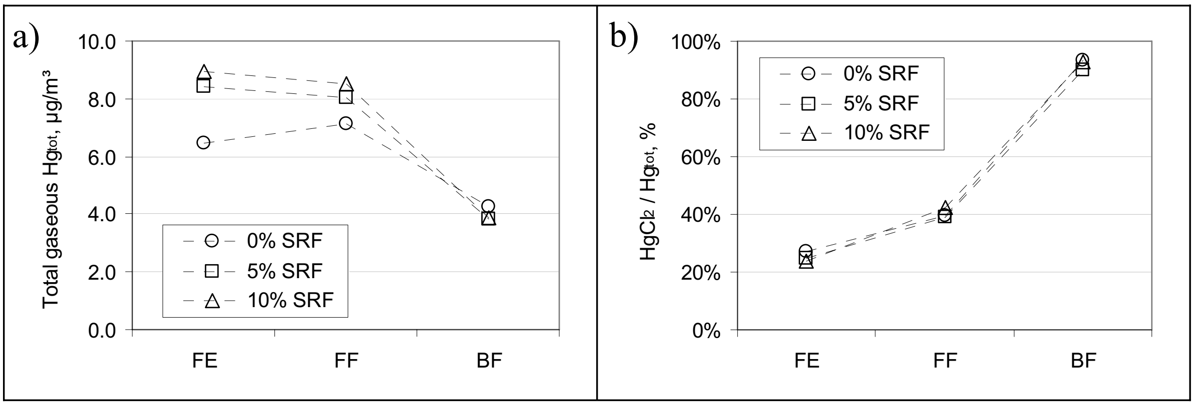


Fig. 5-21: Mercury speciation along the flue gas path a) Total Hg(g), µg/m³ b) $HgCl_2$(g), µg/m³

As discussed beforehand, an oxidation along the flue gas path is seen from about 20% $HgCl_2(g)$ at furnace exit, to 40% ionic mercury in front of the filter. However, a significant increase of ionic mercury is seen across the candle filter to about 100% $HgCl_2(g)$ downstream the filter. Similar results were found by La Marca [60] at a 160 MW_{el} pf- boiler equipped with a bag filter system. In this study, the effect of unburned carbon in ash on mercury removal across the bag filter is studied. However, it is also reported that the ionic mercury share is about 40 - 50% upstream the filter. Across the filter $Hg^0(g)$ decreased significantly, but $HgCl_2(g)$ downstream the filter was almost 100%. The arising question is, whether elemental or ionic mercury adsorbs on the fly ash across the filter. The fact that downstream the filter only ionic mercury is measured suggests that elemental mercury is adsorbed and $HgCl_2(g)$ partially passes the ash filter cake. This is however contrary to literature and laboratory results reported in [61], which means it is generally assumed that ionic mercury adsorbs on fly ash and is removed in ash filtering devices, while elemental mercury does not condense or adsorb on fly ash. Therefore, the following simplified mechanism is suggested according to Fig. 5-22:

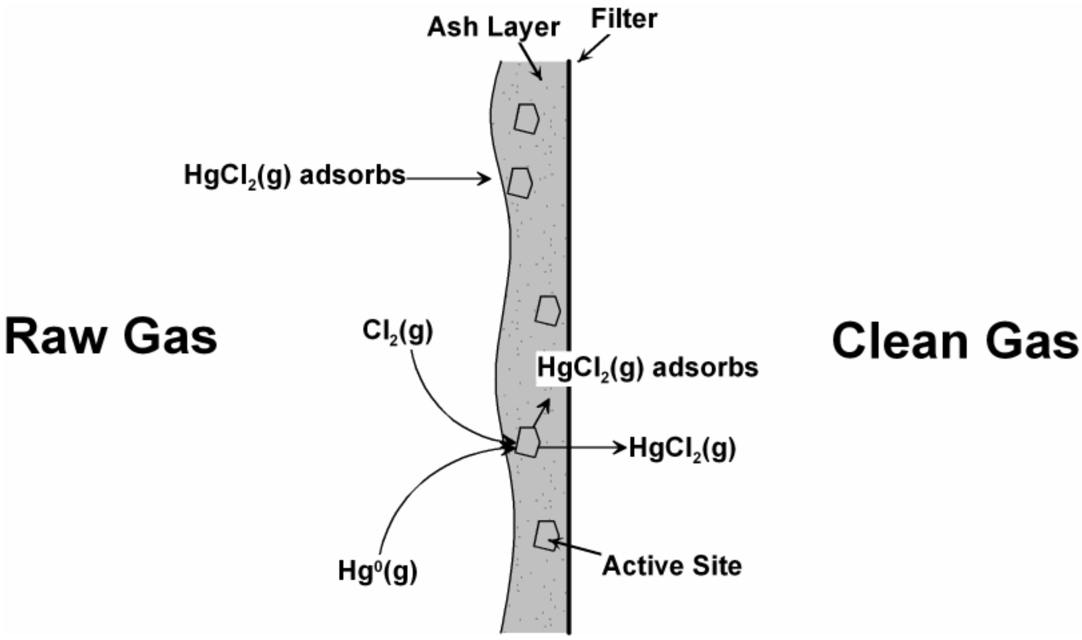


Fig. 5-22: Assumed transformation of $Hg^0(g)$ passing the filter cake of the candle filter system

Upstream the filter the total mercury concentration is made up of already particle bound mercury, mercury chloride and elemental mercury. Hg(p) is then separated with the fly ash when the particle is added to the filter cake. $HgCl_2(g)$ adsorbs on the fly ash on its way through the filter cake and does not or only to a small extent reach the clean gas side of the filter. For $Hg^0(g)$ it is assumed that it

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[60] C. LA MARCA, A. BIANCHI, C. CIONI, S. MALLOGGI: "Mercury Oxidation and Removal in the gas-cleaning system of bituminous-coal fired unit", Proceedings of Air Quality V, Arlington, USA, (2005)

[61] H. THORWARTH, V. STACK-LARA, S. UNTERBERGER, G. SCHEFFKNECHT: „The Influence of Fly Ash Constituents on Mercury Speciation", Proceedings of Air Quality V, Arlington, USA, (2005)

gets oxidised across the filter cake. For the oxidation reaction there are generally several reactions possible as summarised by Hocquel [62], while the most important reactions are with HCl(g), Cl_2(g) and Cl^-(g).

$$Hg^0(g) + 2HCl(g) + \frac{1}{2}O_2 \leftrightarrow HgCl_2(g) + H_2O(g) \qquad (5.4)$$

$$Hg^0(g) + Cl_2(g) \leftrightarrow HgCl_2(g) \qquad (5.5)$$

$$Hg^0(g) + 2Cl^- \leftrightarrow HgCl_2(g) \qquad (5.6)$$

After the oxidation step in the filter cake, one part of the now oxidised mercury adsorbs on the fly ash and one part reaches the clean gas side of the filter.

To summarise the mercury measurement, it was found that co-combustion increases the amount of ash bound mercury for the investigated lignite. A significant influence of residual carbon on ash bound mercury was identified for the investigated bituminous coal. Furthermore, the trials indicated that no or only insignificant amounts of Hg^0(g) could be determined behind the fly ash filter system. Referring to other investigations, it is not likely that Hg^0(g) absorbs on the fly ash. Therefore, it is assumed that Hg^0(g) gets oxidised across the filter cake. Furthermore, the co-combustion trials showed that the concentration of ionic mercury $HgCl_2$(g) was not directly linked to the chlorine concentration in the gas phase. No or only insignificant amounts of gaseous elemental mercury Hg^0(g) could be determined behind the fly ash filter system, while residual $HgCl_2$(g) concentrations do not differ significantly. Therefore, it can be stated that co-combustion of the investigated SRF qualities - in combination with the base coals - does not increase the overall mercury emissions of the process.

b) Behaviour of Cd, Zn, Ni, Cu, Pb

Due to a higher concentration of zinc, lead, and copper in the investigated SRF, an enrichment of these elements can be expected in the mixed fly ash from coal and SRF combustion. This expectation shall be investigated based on the results of the experiments at the BTS combustion reactor. Table 5-35 summarises the results for the lignite L/3 during co-combustion with SRF/04:

Table 5-35: Parameter study: Element concentration in the fly ash, mg/kg

Mode / Element	Cd	Zn	Ni	Pb	Cu
Baseline, 0%	1.0	110	40	25	57
SRF/04, 5%	1.1	170	46	42	69
Comparison	→	↑	→	↑	↑
↓ : Decrease → : Comparable ↑ : Increase [ ] : Uncertain					

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[62] M. HOCQUEL, S. UNTERBERGER, K.R.G. HEIN: "Mitverbrennung von Klärschlämmen in Kohlestaubfeuerungen unter Berücksichtigung des Quecksilberanteils bei unterschiedlichen Rauchgasreinigungssystemen", Final project report, IVD, (2002)

As expected, an increase could be determined for zinc, lead, and copper. Considering cadmium and nickel with comparable concentrations both in SRF and coal, no significant increase could be determined.

5.4.3 Comparison and evaluations of results

The behaviour of heavy metals in the UPSWING process and during SRF co-combustion was investigated in detail. Mercury, an element with a high toxic potential, was given special care during the experimental trials. Considering the UPSWING process, the question in focus was to what extent the conditions found in the UPSWING process affect the behaviour of heavy metals. In this context, it was determined that the additional water input has the largest effect on mercury speciation, shifting the equilibrium towards elemental $Hg^0(g)$. Considering the other investigated elements Cd, Zn, Ni, Pb, and Cu, additional chlorine seems to have the largest effects. The formation of Me_xCl_y obviously leads to lower amounts found in the ash. Nevertheless, no increased emissions are expected as Me_xCl_y should be removed in the FGD system.

Considering direct SRF co-combustion, the fate of waste-borne pollutants was investigated in detail. Contrary to the UPSWING process, pollutants are brought forward into the power plant combustion process, although the mercury content of the investigated SRF is comparable to those of the base coals. Consequently, increased emissions cannot be expected, and this (positive) aspect could be verified. Furthermore, no elemental $Hg^0(g)$ could be determined behind the candle filter system of the test facility. It is therefore assumed that a considerable part of the elemental mercury gets oxidised across the filter cake. Furthermore, a maximum of 10-12% of the mercury input was absorbed by the fly ash, which is slightly higher compared to the baseline test. A further important aspect found was that the concentration of ionic mercury $HgCl_2(g)$ was not directly linked to the chlorine concentration in the gas phase. Considering the other investigated elements Cd, Zn, Ni, Pb, and Cu, a significant enrichment of elements found in high concentration in the SRF (Zn, Pb, Cu) could be determined.

5.5 Fly ash quality and utilisation

The final experimental chapter discusses the influence of both processes on residue quality, with the focus on fly ash quality and its further utilisation. Due to the almost complete removal of fly ash from the waste flue gases, no significant influence is expected for the UPSWING process. Moreover, this aspect is emphasised as on one of the main benefits in comparison to direct co-combustion, as waste-borne pollutants and other components may change the properties or the composition of the ash.

The importance of further unrestricted fly ash utilisation should be reviewed by an overview of the current situation in Germany: In hard coal fired power plants, usually 80 to 90% of the total fuel ash is removed as fly ash via the ash removal system, typically designed as ESP for large-scale power plants. For lignite combustion approx. 80% of the fuel ash is removed as fly ash and 20% as bottom ash. Both fractions are usually mixed. Table 5-36 shows the produced amounts of fly ashes from lignite and hard coal firings in Germany (2001):

Table 5-36: Fly ash production in Germany [63]

Fly ashes from	Year	Production, Mg
Hard coal combustion	2001	4,000,000
Lignite combustion	2001	7,500,000

The amount of lignite fly ashes is twice the amount of hard coal fly ashes. The common utilisation of both types of ashes is different: Fly ashes from hard coal are mainly used in cement and concrete industry, whereas fly ashes from lignite combustion are used as backfilling materials in open cast mines. Both ways of utilisation are directly connected with the ash properties as shown in Table 5-37:

Table 5-37: Typical fly ash composition from lignite- and hard coal fired power plants [64]

Oxide	Unit	Hard coal	Lignite
SiO_2	%	40 - 55	20 – 70
Al_2O_3	%	23 - 35	1 – 15
Fe_2O_3	%	4 - 17	1.5 – 70
CaO	%	1 - 8	8 – 40
MgO	%	0.8 - 4.8	0.5 - 7
K_2O	%	1.5 - 5.5	0.1 - 1.5
Na_2O	%	0.1 - 3.5	0.1 - 4
SO_3	%	0.1 - 2	1.5 - 15
TiO_2	%	0.5 - 1.3	0.1 - 1

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[63] S. KÖSTER: „Relevante Abfallströme für eine Immobilisierung“, Aachen, (2003)

[64] S. PAULY: „Chemie und Biologie der Altlasten“, Fachgruppe Wasserchemie in der Gesellschaft Deutscher Chemiker, (1997)

The values given in the table indicate larger ranges for fly ash components from lignite combustion. Due to this the utilisation in the cement and concrete industry is not common practise as a more or less constant product quality is required. Another factor might be the brown colour of lignite ashes, not fulfilling aesthetic requirements. Nevertheless, the utilisation of lignite ashes is allowed. Concerning fly ashes from hard coal combustion Table 5-38 gives a brief overview of the common utilisation paths in Germany:

Table 5-38: Common utilisation of fly ashes from hard coal combustion [65]

Field of application	Fraction, %
Concrete/mortar	70.1
Mining industry, backfilling material	12.4
Cement industry	10.2
Road construction, etc.	6.6
Brick production	0.7

As the utilisation of both fly ashes from lignite and hard coal combustion are different, applicable legal regulations and limits differ as well.

a) Fly ash from hard coal combustion

As seen in Table 5-38, fly ashes from hard coal combustion are used to a percentage of approx. 70% in the concrete industry. If they are utilised this way, they are within the scope of the European Standard EN-450 (and subsequent norms) on the utilisation of fly ashes in concrete. The most important requirements are summarised in Appendix D-1. According to this standard fly ashes are defined as additives. They are used either to improve or achieve specific properties of the concrete. Two types are defined in the standard, which are inactive additives (Type I) and hydraulic additives (Type II). It should be mentioned that the EN-450 defines no limits in terms of heavy metals.

Considering the fate of heavy metals in concrete originating from the utilisation of fly ash, a possible environmental hazard could be leaching by water, e.g. rain or ground water. Furthermore, a question would be if leaching effects could be enforced, if a construction is subjected to chemical stress like carbonation or salt attack. Leaching effects can also take place at concrete granulate, e.g. on recycling centres or when used in road construction. In this context, leaching tests with water soaked through concrete granulate showed the following results: At the beginning the concentration of solved substances is rather high as they are washed from the surfaces. With passing time, the concentrations decrease rapidly. If the surfaces are cleared, the concentrations are almost zero [66]. Further tests showed that no dependency could be determined between the

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[65] F. SOMMERFELD, G. SCHWEDT: „Vergleich ausgewählter Elutionsverfahren zur Beurteilung der Mobilität von Metallen“, Acta Hydrochemie/Hydrobiologie 24, (1996)

[66] HOLCIM AG: „Zement und Umwelt: eine nachhaltige Bindung“, Edition Holcim AG, Herausgeber: Holcim AG, Siewerdtstrasse 10, CH-8050 Zürich, (2001)

amount of heavy metals in the concrete and the leached concentrations. In most cases, the concentrations of heavy metals were below the respective determination limits. Tests under extreme conditions (high acid concentrations, granulated or pulverised samples) showed that only a few percent of the total element concentration of the samples could be leached out.

Following the above-mentioned findings, the use of fly ashes with slightly increased heavy metal content, e.g. from SRF or municipal sewage sludge co-combustion, should impose no significant environmental risk. Nevertheless, fly ashes from co-combustion processes have to conform to the requirements defined in EN-450 in all relevant aspects. An official approval can be applied for, and it has to be proved that no significant changes of the fly ash properties occur by changed process conditions in the power plant. Furthermore, the environmental impact of the fly ashes and the produced concrete has to be assessed and evaluated against fly ashes from 100% coal combustion.

A further option for fly ash utilisation not yet discussed is the cement industry. Approximately 10% of the fly ashes from hard coal combustion are utilised this way. No legal regulations or limits exist; the utilisation depends on an agreement between the producer and the cement industry. Contrary to this procedure, Switzerland has regulations defining limits for fly ashes. These are summarised in the so-called BUWAL list [67].

b) Fly ashes from lignite combustion

The situation for fly ashes from lignite combustion is different. As mentioned above, they are commonly used as backfilling material in open cast mining. No special regulations or limits are defined for fly ashes in this context; therefore the *Versatzverordnung (VersatzV)* has to be considered in Germany. The relevant limits for inorganic substances, acquired in leaching tests, are summarised in Appendix D-1. Fly ashes from co-combustion processes have to keep these values, too. If the produced fly ashes exceed the given limits, the material would be classified according to the *Abfallablagerungsverordnung (AbfAblV)*, although a deposition of fly ashes as wastes is of course no economical option. The limits given in the *AbfAblV* are annexed in Appendix D-1 as well.

5.5.1 Influence of waste flue gas integration on fly ash quality

The partial flue gas treatment system of the UPSWING process includes almost complete fly ash removal prior to injection into the power plant. This aspect is a major benefit of the entire process as the contamination of power plant ashes by waste-borne particulate matter is basically omitted. Nevertheless, fly ash properties might be altered by gaseous contaminants such as HCl or SO_2. A clear statement towards this aspect, based on the experiments with artificial waste flue gases, is not possible. In order to evaluate this aspect, an idea might be to expose fly ashes sampled in power plants with partially-cleaned waste flue gases from a waste incineration plant.

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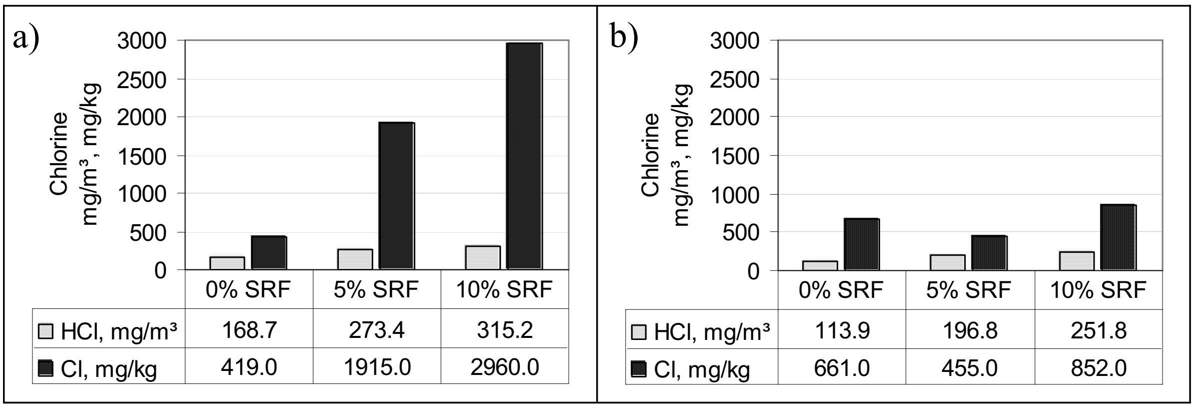
[67] D. TRABER, F. JACOBS, U. MÄDER, U. EGGENBERGER: „Einsatz von Sekundärstoffen im Beton: Technische und ökologische Anforderungen“, Betonwerk und Fertigteil-Technik, Vol. 66, S. 76-84, (2000)

5.5.2 Deterioration of power plant residues during SRF co-combustion

The (potential) deterioration of power plant residues during SRF co-combustion should be discussed based on the fly ashes acquired during the trials at the semi-technical combustion facility KSVA (refer to chapter 4.2). Experiments were performed using two different base fuels (German bituminous coals B/2 and B/3) and one SRF quality (SRF/PRR) in two different thermal shares (5/10%). The experimental settings are summarised in Appendix C-2.

An important factor is the high chlorine content of the SRF material (approx. 1% in the raw material). Chlorine in fly ashes utilised as additives in concrete may attack embedded steel by corrosive attacks. As a consequence, the chlorine content in fly ash is limited to a maximum of 0.1%. Therefore it has to be questioned, if and to what extent the additional chlorine from the SRF influences the quality of the fly ash. Fig. 5-23 summarises the results for chlorine:



a)

	0% SRF	5% SRF	10% SRF
□ HCl, mg/m³	168.7	273.4	315.2
■ Cl, mg/kg	419.0	1915.0	2960.0

b)

	0% SRF	5% SRF	10% SRF
□ HCl, mg/m³	113.9	196.8	251.8
■ Cl, mg/kg	661.0	455.0	852.0

Fig. 5-23: Results for gaseous HCl and ash-bound chlorine a) SRF/PRR.B/2 b) SRF/PRR.B/3

As expected, SRF co-combustion leads to a significant increase of gaseous chlorine, measured as hydrogen chloride (HCl, refer to chapter 5.2ff). Furthermore, an increase of ash-bound chloride can be determined. Differences between both coals are obvious: For the bituminous coal B/2, chloride increases from 420 mg/kg (baseline) to approx. 3000 mg/kg (0.3%) for a co-combusted share of 10%. For bituminous coal B/3, the effect is much lower, ranging from 450 to 850 mg/kg. Reasons explaining this difference can be found in the ash constituents of both coals, especially in calcium oxide (CaO), which is expected to absorb the additional chlorine freight ($CaO + 2HCl \leftrightarrow CaCl_2 + H_2O$). The bituminous coal B/3 has a CaO content of $3.4\%_{ash}$, the bituminous coal B/2 of $10.5\%_{ash}$. In addition, the total ash content of B/2 is higher (8.3% vs. 5.4%, see Appendix A-1). As a consequence, more calcium is available to absorb the additional waste-borne chlorine. Considering the trials for the bituminous coal B/2, limits for chlorine as defined by EN-450 (and subsequent norms) are exceeded during co-combustion.

In the context of pollutant absorption by the fly ash, another important ash component in terms of further utilisation is sulphur. Sulphur, expected to form anhydrides (SO_3), is limited to < 3%. Fig. 5-24 shows the ash bound SO_3 concentration as well as the calculated recovery rate:

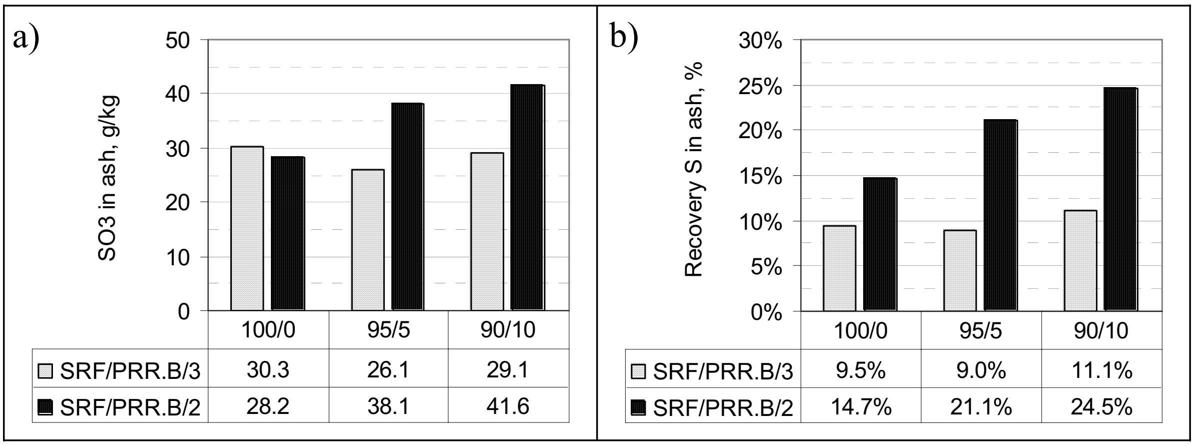


Fig. 5-24: a) SO_3 in ash, g/kg b) Recovery of sulphur in ash, %

Comparable to the results found for chlorine, an increased amount of sulphur is absorbed by the fly ash for the bituminous coal B/2. The balance indicates that approx. 25% of the sulphur is bound to the fly ash for a thermal share of 10%, during baseline trials only 15%. B/3 shows only a slight increase. Aside from chlorine and sulphur, other relevant fly ash properties were investigated as well, especially loss on ignition (LOI), sum of silicon, aluminium, and iron, alkali content and free calcium oxide. The requirements according to EN-450 are summarised in Appendix D-1. Analytical data were obtained by standard XRF analysis of the co-combustion ashes. Table 5-39 summarises the results for the system **SRF/PRR.B/2** for baseline coal combustion (100/0) and the replacement of 5% (95/5) respectively 10% (90/10) of the thermal heat input:

Table 5-39: Characterisation of fly ash properties according to EN-450 for system SRF/PRR.B/2

Parameter	100/0	95/5	90/10	Requirement
LOI, %	4.2	-	2.0	see below *
$SiO_2 + Al_2O_3 + Fe_2O_3$, %	79.8	77.3	74.6	> 70%
Chloride, %	0.04	0.19	0.29	< 0.1%
SO_3, %	2.8	3.8	4.2	< 3% **
Free calcium oxide, %	1.22	-	1.28	< 1%
Total alkali content, %	2.4	3.0	2.5	< 5%
* LOI grade A (< 5%), B (2-7%), C (4-9%) ** calculated from ultimate analysis				

As discussed above, co-combustion of the present SRF leads to an increased chlorine and sulphur content of the fly ashes. Both parameters exceed the limits defined in EN-450 (and subsequent norms) during co-combustion. Apparently, fuel burnout (LOI) is not influenced by co-combustion, which is a positive result. A slight decrease of $SiO_2+Al_2O_3+Fe_2O_3$ could be observed during co-combustion, resulting from lower concentration of these elements in the SRF. Nevertheless, the requirements would be still fulfilled for the co-combustion ashes. No significant difference could be observed for the total alkali content. The acceptable limit for free calcium oxide of 1% is exceeded even during baseline coal combustion.

Table 5-40 summarises the results for the system **SRF/PRR.B/3** for baseline coal combustion (100/0) and the replacement of 5% (95/5) respectively 10% (90/10) of the thermal heat input:

Table 5-40: Characterisation of fly ash properties according to EN-450 for system SRF/PRR.B/3

Parameter	100/0	95/5	90/10	Requirement
LOI, %	15.0	13.9	13.2	see below *
$SiO_2 + Al_2O_3 + Fe_2O_3$, %	77.9	81.3	80.9	> 70%
Chloride, %	0.07	0.05	0.09	< 0.1%
SO_3, %	3.0	2.6	2.9	< 3% **
Free calcium oxide, %	0.41	-	0.33	< 1%
Total alkali content, %	3.1	3.7	3.3	< 5%
* LOI grade A (< 5%), B (2-7%), C (4-9%) ** calculated from ultimate analysis				

Comparing the results for LOI with bituminous coal B/3, significant higher levels can be observed, although an increased co-combusted share of SRF seems to improve final fuel burnout. Despite of this result, no major influence of SRF co-combustion on the ash properties could be observed for this specific coal. The absorption capacities of the coal ash seem to be completely different to B/2 and shall be characterised as “neutral” at this point. Contrary to bituminous coal B/2, co-combustion of B/3 with this type of SRF seems to be possible in terms of further ash utilisation.

5.5.3 Evaluation of fly ash quality according DIN/EN 450

With the focus on SRF co-combustion, the requirements found in European Standard EN-450 on the utilisation of fly ashes in concrete were applied on ashes obtained during semi-technical co-combustion trials based on two different bituminous coals and one SRF quality. Based on the experimental results, the following statements can be made:

- Low- to medium volatile coals may benefit from combustion of SRF due to its significant volatile content.
- Calcium-rich coals tend to absorb a significant amount of sulphur and chlorine. Comparable results were found during the trials at the BTS combustion reactor described in chapter 5.2.2.
- No significant influence of SRF co-combustion could be determined for the amount of free calcium oxide and total alkali content for the investigated coals in comparison to baseline 100% coal combustion.

Generally, the composition and properties of the mixed co-combustion ashes depends on both ash composition of coal and SRF. As shown above, certain limits were partly exceeded for the bituminous coal B/3. In order to avoid unwanted effects on fly ash quality, it is therefore strongly recommended to evaluate possible effects on fly ash quality in dedicated trials before large-scale application.

6 Evaluation of the full-scale process by numerical simulation

Flue gas integration is one of the most important aspects of the UPSWING process. Although partly cleaned, the flue gases from the waste incinerator unit still carry a significant pollutant load, which has to be either reduced (in terms of nitrogen and sulphur oxides) or destroyed (in terms of organic pollutants) in the power plant process. The whole complex was intensively discussed in the previous chapter. During the course of the work, it soon became obvious that it would not be possible to get a final statement upon operational consequences of flue gas integration for the full-scale power plant process by means of lab- to semi-technical trials alone. This does not mean that the test facilities described in chapter 4 are not suitable to investigate specific aspects such as NO_x reduction potential or PCDD/F destruction, but they do not picture the entire process sufficiently. To overcome this deficit, it was decided to investigate the full-scale application of the UPSWING concept by numerical simulation. Hence, a full-scale lignite fired boiler was modelled using the 3D-CFD code AIOLOS, developed by IVD, which allows the simulation and evaluation of different operational settings such as variable waste flue gas properties or the specific injection location into the boiler.

6.1 Basic approach

It was discussed in the previous chapter that the injection of cold and wet flue gases has significant influence on the power plant process, possibly reducing the flame temperature and worsening coal burnout. From the experimental investigations performed it is known that the specific injection location has consequences on the emission behaviour, especially in terms of nitrogen oxides (NO_x). As a consequence, the entire complex has to be investigated more closely. Considering flue gas integration in large-scale perspective, four major alternatives have been identified as technically feasible:

(1) **Vapour burners**: In case of lignite-fired power plants, the raw lignite is dried and pulverised in beater-wheel mills, commonly one mill per burner. If the mill is equipped with a vapour separation system, dried pulverised coal and water vapour are separated behind the mill to increase coal concentration at the main coal burners. Due to the specification of the waste flue gases to be injected into the power plant process (wet, cold, low-oxygen content), the injection via the vapour burners together with the coal vapours proposes to be a suitable option with minimal effects on the entire system.

(2) **Burner air system**: The injection via the burner air system can be considered as well. In this case, mixing of the waste flue gases with the combustion air would be necessary. To allow this, the pressure of the waste flue gases would have to be increased by an appropriate fan to meet the pressure of the secondary air system. From an operational

point of view this option cannot be recommended as this solution dramatically increases the risk of leakage and contamination.

(3) **Additional injection nozzles**: The application of additional nozzles above the main burner belt was considered as well. Advantageous would be that the coal flame would not (or only to a minor extent) be influenced by the flue gases. Furthermore, the pressure must not be increased prior to injection into the furnace.

(4) **Ash hopper**: The ash hopper, or the lower part of the boiler, is another favourable option for flue gas injection. In this case, the flue gases would be injected below the main combustion zone. Advantages of this solution would be once again the low pressure and a minimum requirement of moving parts such as compensators.

Following the basic considerations made above, options 1, 3, and 4 were investigated in detail. In this context, an important subject to modelling is the quality of the numerical grid itself. For the planned investigations, a numerical grid with approximately 1.8 million calculated cells was created. The memory demand of approx. 3GB required to run the simulation on a supercomputer in order to gain the results in an acceptable timeframe. This was achieved by using one node, comprising of eight processors, on the NEC-SX8 platform at the HLRS (High Performance Computing Center) Stuttgart. Fig. 6-1 shows the whole grid and the zoomed-in burner domain:

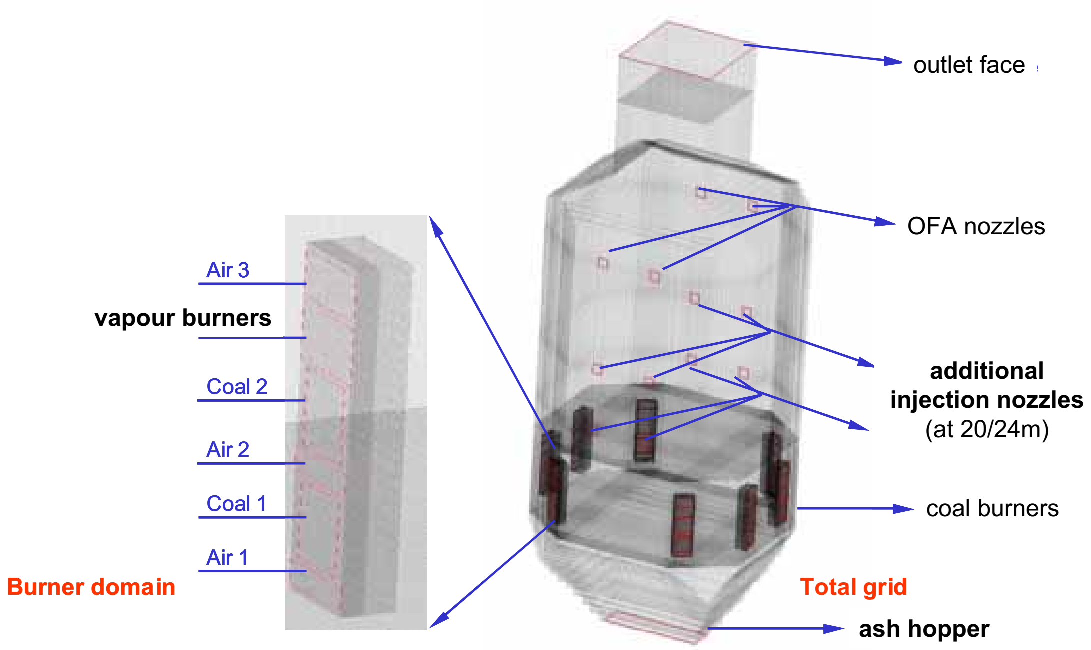


Fig. 6-1: Numerical grid of the lignite-fired boiler

The figure shows the complexity of the grid. As mentioned above, the injection locations taken into consideration were the vapour burners (vapours), the ash hopper (hopper inlet), and additional injection nozzles. The retrofitted nozzles were designed at 20.0m and 24.0m, respectively, located between the vapour and over-fire air nozzles (OFA).

A further important aspect is the quality of the models used. A common approach in numerical simulation is to validate the results using data obtained during measurement campaigns. In case of the UPSWING concept, which is not yet realised, this option is not available. To overcome this deficit, it was decided to use the experimental data obtained in lab- to semi-technical trials to improve and validate the reliability of the combustion and NO_x models implemented in the CFD code. This approach includes the modelling of the combustion reactor BTS (refer to chapter 4.1) and the adaptation of the existing NO and combustion models for a specific lignite, which will be briefly discussed in chapter 6.2.
The next step includes the implementation of the adapted models in AIOLOS and their application for the large-scale case, whereas the different operational cases were simulated and evaluated by means of a dedicated parameter study in order to identify the best-suited technical solution. This is subject of chapter 6.3. In extension to this first set of simulations, chapter 6.4 discusses the evaluation of favourable process settings by an extended parameter study.

6.2 Model adaptation and validation

AIOLOS is based on a conservative finite-volume formulation, using the SIMPLEC- or SIMPLE-method for velocity-pressure coupling, standard k-ε-model or differential Reynolds stress model for turbulence. The program includes a four-step-reaction scheme, two for heterogeneous reactions of pyrolysis and char combustion and two for the gas phase reactions. Radiative heat transfer is calculated either by a discrete-ordinates-method or five other different radiation models [68]. More detailed information about the code AIOLOS can be drawn from Schnell [69] and Förtsch [70].

For the simulation of the combustion reactor BTS, a 2D numerical grid was generated, formulated in cylindrical coordinates. In a first step, simulations were carried out in order to correctly depict the coal combustion properties at the furnace exit. Therefore, for each experimental setup in case of changing boundary conditions, e.g. air-to-fuel ratio, the modelled particle size distribution of the coal was adapted to guarantee a complete coal/char burnout. Secondary air was fine-tuned to correctly predict the measured O_2/CO_2-concentrations at the furnace exit. In a second step the NO_x model was applied as a post-processor to determine the NO_x formation in the combustor. The distribution of nitrogen in the products of primary pyrolysis (HCN, NH_3, tar-N and char-N) as an initial boundary condition for the NO_x model has a major influence on NO_x prediction.

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[68] H. KNAUS, R. SCHNEIDER, X. HAN, J. STRÖHLE, U. SCHNELL, K.R.G. HEIN: „Comparison of Different Radiation Heat Transfer Models in Coal-Fired Utility Boiler Simulations Using Boundary-Fitted and Cartesian Grids", 4th Int. Conf. on tech. comb. for a clean environ., Lisbon, Portugal, (1997)

[69] U. SCHNELL: "Numerical Modelling of Solid Fuel Combustion Processes Using Advanced CFD-Based Simulation Tools", Progress in Computational Fluid Dynamics Vol. 1, No.4, pp. 208-218, (2001)

[70] D. FÖRTSCH: "A Kinetic Model of Pulverised Coal Combustion for Computational Fluid Dynamics", PhD thesis, University Stuttgart, (2003)

Hence, variations of these parameters have been used to adapt the predictions of the NO_x model for every experimental set-up to the measured values, in order to gain model parameters for the later application to the large-scale power plant. Aside from baseline coal combustion, two operational settings have been investigated in detail, viz.

> *Case A: Additional injection of flue gases*
> Waste flue gases were injected via the secondary air duct of the burner [A.1], or injected at 0.9 m distance from the burner below the flame [A.2], respectively. Combustion air is not reduced. As a consequence, the overall oxygen content in the combustion chamber increases from 3.5% increases to 4.1%.
>
> *Case B: Substitution of combustion air*
> Waste flue gases were injected via the secondary air duct of the burner [B.1], or injected at 0.9 m distance from the burner below the flame [B.2], respectively. Combustion air was partly reduced - or substituted - using the remaining 10% oxygen in the incinerator flue gases as oxidising medium for coal combustion, and the oxygen content in the combustion chamber remained constant (3.5%).

The simulation results show a quite good correspondence between predicted and measured results for both baseline coal combustion (see Fig. 6-2a) and flue gas injection (see Fig. 6-2b). Even the different injection locations can be described well enough.

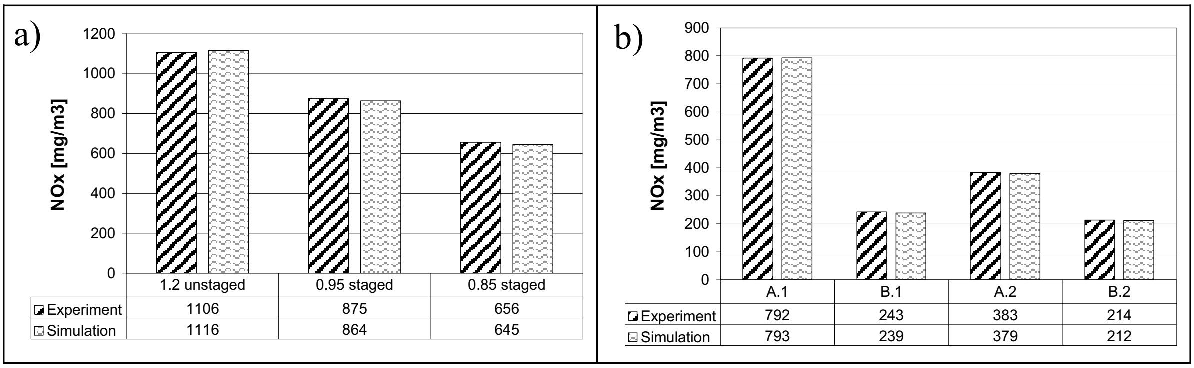


Fig. 6-2: a) Flue gas residence time 1s b) Flue gas residence time 2s

It can be summarised that the implementation of adapted model parameters leads to favourable results for the experiments performed at the test facility. With the utilisation of the adapted model parameters in the large-scale boiler simulation, a sufficient reliability can be expected, thus enhancing the quality of the prediction. Hence, the results should be suitable to evaluate and further assess the influence of waste flue gas injection on the full-scale process.

6.3 Evaluation of the best-suited flue gas injection concept

After the implementation of the adapted model parameters, the influence of waste flue gas integration on the power plant process was investigated by large-scale simulation. The simulations were calculated for standard seven-mill operation (n-1). Asides from baseline coal operation (reference case), the injection of waste flue gases via the vapour burners and via additional nozzles was investigated. Table 6-1 summarises the simulated cases:

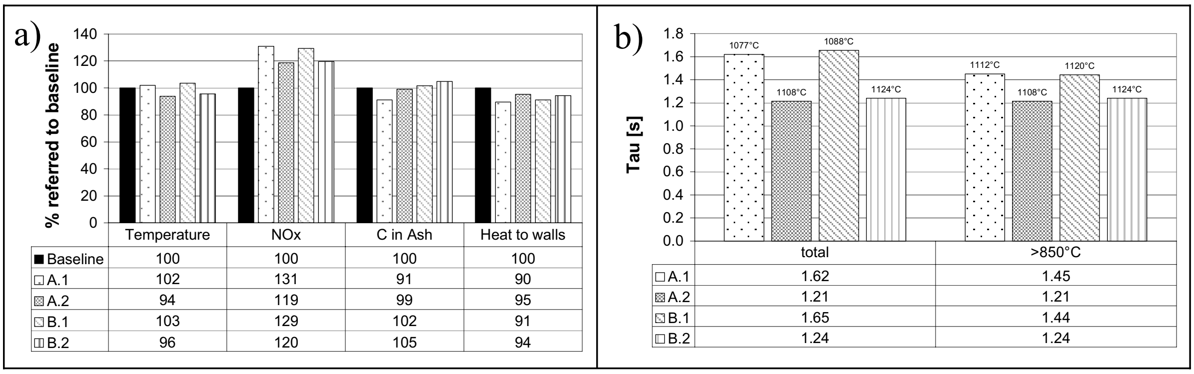
Table 6-1: Characterisation of the simulated cases

Case → Operational mode ↓	**Baseline**	**A.1**	**A.2**	**B.1**	**B.2**
Mills in operation	7 of 8	7 of 8	7 of 8	7 of 8	7 of 8
Injection location of the waste flue gases	-	Vapour burners	Additional nozzles	Vapour burners	Addtional nozzles
Flue gas ratio *	-	10%	10%	10%	10%
Oxygen level of flue gases at furnace exit	-	4.1%	4.1%	3.5%	3.5%
* ratio of waste flue gas to flue gas from coal combustion					

A further relevant parameter - especially in terms of PCDD/F destruction - is the residence time of the waste flue gases in the coal-fired boiler. Generally, the longer this residence time in combination with the high temperature in the furnace, the more efficient should be the destruction of micro-organic pollutants. In order to assess this parameter, the residence time of the waste flue gases from inlet to furnace exit was approximated.

6.3.1 Simulation results

The results of the different cases considering the furnace exit values of temperature, NO_x and fuel burnout (C in ash), but also the radiative and convective heat flux to the furnace walls (heat to walls), are shown in Fig. 6-3a. The results for the individual cases refer to baseline coal combustion. The flue gas residence times correlated to the mean temperatures are given in Fig. 6-3b. The columns on the left display the total residence times of the flue gases from their individual inlets, while the right columns represent the residence times once the gases been have heated up above 850°C.



	Temperature	NOx	C in Ash	Heat to walls
■ Baseline	100	100	100	100
□ A.1	102	131	91	90
▨ A.2	94	119	99	95
▧ B.1	103	129	102	91
◫ B.2	96	120	105	94

	total	>850°C
□ A.1	1.62	1.45
▨ A.2	1.21	1.21
▧ B.1	1.65	1.44
◫ B.2	1.24	1.24

Fig. 6-3: a) Simulation results for the investigated cases b) Flue gas residence times in the furnace

Slight differences can be identified for the furnace exit temperatures. Remarkable are the predicted NO$_x$ emissions, which are significantly higher compared to baseline operation. Considering C in ash, differences can be identified between the two different operational settings, namely additional injection [A] and substitution of combustion air [B], whereas final burnout seems to benefit from additional flue gas injection as a result of the increased oxygen level. The heat flux to the walls is reduced for all investigated cases. The mean residence times of the flue gases in the furnace range between 1.2 and 1.5s for temperatures above 850°C.

6.3.2 Implementation of criteria for process evaluation

A method was sought in order to compare and evaluate the different investigated cases. It was decided to use the parameters investigated above, which are temperature, NO$_x$, C in ash, heat to walls, and residence time, and compare them to baseline coal operation. Following this approach, it is necessary to assess the consequences of the individual parameters on boiler operation:

- **Temperature** (furnace exit): Higher mean temperatures may improve the fuel burnout, but can cause slagging and fouling problems in the superheater region. Lower temperatures delay the fuel burnout. Therefore, temperatures comparable to baseline operation are favoured.
- **NO$_x$ emissions**: An increase of the total NO$_x$ emissions is generally considered negatively.
- **C in ash** (fuel burnout): Increased carbon content at the reactor exit is worse in terms of the overall combustion efficiency. Hence, economical aspects are involved as well.
- **Heat flux to evaporator**: The radiative and convective heat flux to the furnace walls. The lower this heat flux, the more energy remains in the gases and has to be removed in the convective part of the boiler, accordingly. This may cause problems. Consequently, a comparable or higher heat flux is favoured.
- **Residence time**: Longer residence times at higher mean temperatures are considered beneficial to ensure the destruction of micro-organic pollutants.

Based on this assessment, and in order to enable a comparison of the results, ranges were defined for the individual parameters as shown in Table 6-2:

Table 6-2: Criteria for process evaluation

Rank / Criteria	Temperature	NO$_x$ emission / C in ash	Heat flux to evaporator
much better: ++	▨	< -10%	> +10%
better: +	▨	-10% ≤ x <-5%	+5% < x ≤ +10%
comparable: 0	-5% ≤ x ≤ +5%	-5% ≤ x ≤ +5%	-5% ≤ x ≤ +5%
worse: -	-10% ≤ x <-5% +5% > x ≥ +10%	+5% < x ≤ +10%	-10% ≤x < -5%
much worse: - -	< -10% and > +10%	> +10%	< -10%

6.3.3 Evaluation of simulation results

It has to be stated, that all investigated injection concepts have a more or less negative impact on the coal boiler, e.g. in terms of emissions, reduction of the flame temperature, or fuel burnout. This could be expected, as the injection of relatively cold and wet flue gases do not appear beneficial for boiler operation. Despite of this, a favourable injection concept would be one with moderate effects on combustion behaviour and boiler performance. In order to find this concept, the criteria defined in the previous chapter were applied upon the simulation results described in chapter 6.3.1. The results of this assessment are summarised in the following table:

Table 6-3: Evaluation of investigated concepts

Parameter / Case	A.1	A.2	B.1	B.2
Injection location	Vapour burners	Additional nozzles	Vapour burners	Additional nozzles
Oxygen level at furnace exit	4.1%	4.1%	3.5%	3.5%
Temperature	0	-	0	0
Residence time	+	-	+	-
NO_x	- -	- -	- -	- -
C in ash	+	0	0	0
Heat to walls	-	0	-	-

Introducing wet flue gases via the vapour burners as simulated in the variants A.1 and B.1 results in a better burnout of the fuel and a longer residence time of the flue gases, compared to the other cases A.2/B.2, but suffers from a lower heat exchange to the evaporator and higher NO_x emissions. However, all investigated variants produce some 20-30% additional NO_x. Considering the concept with the least effects on combustion behaviour and boiler performance, option A.1, the additional injection of the incinerator flue gases via the vapour burners, is the favourable option. Benefits of this waste flue gas integration concept are favourable fuel burnout performance (lowest C in ash including the baseline case) and a furnace exit temperature comparable to baseline operation.

6.4 Determination of favourable process settings

The previous chapter focused on the best-suited injection location, assuming constant waste flue gas composition. An extended parameter study is therefore required to investigate variable flue gas properties, including different temperatures, water content, and load ratio (the amount of waste flue gas in relation to the flue gases produced from coal). The previous study indicated that the additional injection of waste flue gases via the vapour burners is the favourable option A.1. In order to reduce the amount of necessary simulation runs, injection via additional nozzles (A.2) as well as the operational concept of combustion air substitution were skipped (B.1/B.2). In addition, a not yet discussed possibility was considered: The injection via the hopper inlet, in the following referred to as option C. The investigated cases are summarised in the following table:

Table 6-4: Characterisation of investigated cases

Case → Operational mode ↓	**A.1b**	**A.1c**	**A.1d**	**C.1b**	**C.1c**	**C.1d**
Mills in operation	7 of 8	7 of 8	7 of 8	7 of 8	7 of 8	7 of 8
Injection location of waste flue gases	Vapour burners	Vapour burners	Vapour burners	Hopper inlet	Hopper inlet	Hopper inlet
Flue gas ratio	5%	10%	10%	5%	10%	10%
Flue gas temperature	200°C	400°C	200°C	200°C	400°C	200°C
Water content in flue gas, kg/Nm^3	0.3	0.3	0.0	0.3	0.3	0.0
Substitution of combustion air	no	no	no	no	no	no

Considering the properties of the injected waste flue gases, two temperatures (200/400°C) and two load ratios (5/10%) were investigated. Furthermore, the influence of the additional water in the flue gases was evaluated (0.3 kg/m^3 are expected behind the wet scrubbing process). Comparable to the investigation made in chapter 6.3, the parameter furnace exit temperature, residence time, NO_x emissions, C in ash, and heat to walls were considered and compared with the results acquired in chapter 6.3.1. The results of this extended study are summarised in the following table:

Table 6-5: Evaluation of the investigated cases

Parameter / Case	**A.1b**	**A.1c**	**A.1d**	**C.1b**	**C.1c**	**C.1d**
Injection location of waste flue gases	Vapour burners	Vapour burners	Vapour burners	Ash hopper	Ash hopper	Ash hopper
Temperature	0	0	0	0	0	0
Residence time	+	0	0	+	-	-
NO_x	+	0	+	-	- -	- -
C in ash	0	0	++	++	++	++
Heat to walls	+	0	0	0	- -	- -

Modifying option A.1 by injecting dry flue gases (A.1d) offers additional advantages in comparison to all other investigated cases. Fuel burnout is slightly improved and NO_x emissions are lower. Option A.1b, a reduced flue gas ratio, appears to be beneficial as well. An increased temperature of the flue gases A.1c shows no significant benefits. Considering the hopper cases, C.1b-C1.d, C.1b is superior to the other studied hopper cases and turned out to be an attractive alternative in comparison to the best evaluated vapour burner cases A.1b/A.1d. A main result of the extensive study is obviously the fact that a flue gas ratio of 5% appears to be beneficial, both for hopper and vapour burner injection. This is especially of relevance if the waste flue gases contain a significant amount of water. Nevertheless, a reduction of the flue gas ratio will limit the size of the waste incinerator combined with the power plant.

7 Summary and conclusions

In the previous chapters, the environmental and operational performance of the UPSWING process was evaluated by means of a process comparison with direct SRF co-combustion, and the results will be reviewed and summarised in chapter 7.1. Furthermore, not all operational aspects could be investigated in lab-scale trials. Consequently, these aspects, strongly related to large-scale implementation of the process, will be addressed in chapter 7.2. Finally, an assessment of the economical perspective of the UPSWING process is done in chapter 7.3 by a comparison of the expected costs for thermal waste treatment. The conclusions can be found in chapter 7.4.

7.1 Environmental and operational aspects

The performance of both UPSWING process and direct SRF co-combustion was investigated and compared towards environmental relevant aspects such as process NO_x emissions, fate of hydrogen chlorine, destruction of PCDD/F, and trace element behaviour with the focus on mercury. The investigations followed the idea to compare both UPSWING and direct SRF co-combustion with the standalone power plant process. This was done behind the background of maintaining the (high) standards of conventional municipal solid waste incineration (MSWI).

In terms of NO_x emissions, it was found that both processes have the potential to reduce direct NO_x emissions in comparison to 100% coal combustion. In case of direct SRF co-combustion, the benefits seem to derive from higher share of volatile matter in the material. Lower specific process emissions could be determined as a function of the co-combusted share of SRF. Considering the UPSWING process, it became obvious that the location of the flue gas injection as well as maintaining the burner stochiometry is crucial in terms of lowest process emissions. Furthermore, it was found that the reduction of the additional NO_x load of the injected waste flue gases is generally possible. This assumption derives from the fact that no significant influence of an additional NO_x load was determined for any of the investigated coals, especially if primary NO_x reduction measures are applied. Significant differences were found between the investigated coals and their sensitivity upon waste flue gas injection. From this point, the necessity of well-controlled process conditions has to be emphasised.

In terms of the co-combustion process, the expected increase of gaseous chlorine (HCl) as a result of higher chlorine content in the SRF materials could be verified. Nevertheless, the HCl concentration alone allows no assessment of a potential increased corrosion risk. Considering the amount of chlorine bound to the ashes, significant differences could be determined between the investigated bituminous coal and the lignite. The increased chlorine content in the lignite ashes may be taken as an indicator for an increased corrosion risk. For the

UPSWING process it can be stated that the partial flue gas treatment concept consequently limits the introduction of waste-borne chlorine into the power plant process. Furthermore, no waste-borne ashes are introduced into the power plant process as a result of almost complete ash (and dust) removal prior to flue gas injection. Contrary to SRF co-combustion, no unwanted introduction of critical ash constituents such as alkali chlorides takes place. Consequently, the risk of increased boiler corrosion is significantly lower for the UPSWING concept.

Considering the destruction of PCDD/F, the combustion experiments with spiked coal documented a destruction yield larger than 99.9% for toxic organic compounds. Experiments were performed at two test facilities (pulverised fuel fired and bubbling/circulating fluidised bed). No difference in the results obtained during the reference tests - those without spikes and those when the spiked coal was burnt - could be found. Based on these results, it can be concluded that the injection of PCDD/F containing flue gases from waste incineration into a power plant causes no environmental risk. Considering SRF co-combustion, the results acquired during large-scale co-combustion trials at a lignite-fired coal boiler indicated no increase of PCDD/F emissions. Furthermore, no significant influence on other process specific parameters such as dust or unburnt carbon could be determined.

Considering trace elements such as heavy metals, their behaviour in the UPSWING process and during SRF co-combustion was investigated in detail. Mercury, an element with a high toxic potential, was given special care during the experimental trials. Considering the UPSWING process, the question in focus was to what extend the conditions found in the UPSWING process affects mercury. In this context, it was determined that the additional water input has the largest effect on mercury speciation, shifting the equilibrium towards elemental $Hg^0(g)$. Contrary to the UPSWING concept, pollutants are brought forward into the power plant combustion process during SRF co-combustion, although the mercury content of the investigated SRF materials was comparable to those of the base coals. Increased emissions cannot be expected, and this (positive) aspect could be verified. Furthermore, no elemental $Hg^0(g)$ could be determined behind the ash filter system of the test facility. It is therefore assumed that a considerable part of the elemental mercury gets oxidised across the filter cake. A further important aspect found was that the concentration of ionic mercury $HgCl_2(g)$ was not directly linked to the chlorine concentration in the gas phase.

7.2 Further operational aspects

From a technical point of view, it should be clear that not all aspects related to the large-scale implementation of both UPSWING and direct SRF co-combustion could be evaluated by lab- to semi-technical investigations. Hence, the following chapter will discuss several aspects important for both processes, including process integration, heat recovery, flexibility of plant operation, and emergency situations.

7.2.1 Waste flue gas integration / SRF handling and feeding

Considering the UPSWING process, coupling of the waste incinerator to the coal-boiler on the flue gas side will present only few problems. The flue gases are at relatively low temperatures after the wet-scrubbing stage, less than 200ºC, and the flue gas volumes from the incinerator are manageable, and the routing of the ductwork to the coal boiler should not represent significant difficulties. An adequate insulation has to be considered to omit condensation in the ductwork. In chapter 6 the injection via the vapour burners of a lignite-fired boiler was determined as one preferable option. However, as moving parts are present, there is a risk of leaking and contamination of the surrounding area. From this point of view, another option for waste flue gas injection to the pulverised coal boiler could be the furnace bottom ash hopper. The bottom ash hopper is fixed and routing of the ducts should impose no significant problems. The introduction of the flue gases to the coal-fired boiler will have an impact on the furnace and the convective section heat transfer, and the overall efficiency of the coal boiler. It is also clear that this may be one of the factors which determine the maximum allowable flue gas flow rates to the boiler. Considering the lignite-fired boiler investigated in chapter 6, a flue gas ratio of 5% seems to be beneficial. However, this aspect will be highly site-specific.

Regarding the direct SRF co-combustion concept, delivery, storage, and feeding of the material have to be considered. There is a significant demand of additional space, and the transport to the combustor may cause further problems. In this context, spoiling should be mentioned, and the requirement for a suitable weather and fire protection. Concerning the injection into the combustor, mixing of coal and SRF prior to the coal milling and drying process appears to be suitable in case of lignite fired power plants, usually equipped with beater-wheel mills. In hard coal fired plants, both coal and SRF should be kept separately, as the hard coal mills such as bowl type mills are not suitable for the fibrous and ductile SRF. Possible injection locations would include the main coal duct behind the mill(s), the burner air system or separate burners. The utilisation of a separate mill could be favoured if the SRF material is not “furnace-ready”. In any case, a homogenous distribution of the fuel in the furnace should be targeted in order to omit local concentration peaks of potentially harmful substances.

7.2.2 Heat recovery

Considering the UPSWING concept, the waste incinerator will (usually) generate steam at approx. 400ºC and 40 bar. The most likely options for integration of the steam from the incinerator into an existing power plant are into the cold reheater/feedwater system of the coal boiler, or to a low pressure ring main of the coal power plant, with pressure reduction through a back pressure turbine, if appropriate. There will be a limit on the quantity of steam from the incinerator that can be accepted by the coal boiler for feedwater heating or into the ring main, and this may be an additional limiting factor on the relative throughputs of the waste incinerator and the coal boiler. According to Livingston, the application of the UPSWING concept is, in general, very much easier on the flue gas side than on the steam side, and that the flue gas side is subject to fewer site-specific issues [70]. The problems on the steam side are basically a result of the fact that most large power stations are reasonably well optimised and that it is only possible to find valuable use for a limited amount of additional steam, particularly when generated by the incinerator at fairly modest steam conditions. In this retrofit situation, no increased efficiency in waste-to-energy conversion can be expected compared to the standalone MSWI process. However, this situation would be completely different for newly commissioned plants. If both, waste incinerator and power plant, are designed and optimised for combination, significant benefits in overall process efficiency can be expected. Concerning the SRF co-combustion concept, the heat recovery will take place in the boiler system of the power plant, whereas the net efficiency should be comparable to those of baseline coal combustion.

7.2.3 Flexibility of plant operation

Another important aspect is the flexibility of plant operation. Restrictions for the power plant process can be expected if a waste incinerator is connected both on the steam and on the flue gas side. Furthermore, additional efforts in control and regulation including the necessary auxiliary equipment have to be considered. To increase the overall process flexibility, the waste incinerator should be connected to more than one coal boiler on the steam side as well as on the flue gas side. Nevertheless, the direct SRF co-combustion concept appears to be more flexible in this context. The additional boiler fuel can be switched on and off, although the storage situation on the site has to be respected, accordingly. A further aspect which concerns both concepts is the availability of the power plant. The usual operating time of a waste incinerator ranges between 7500 to 8000 h/a, and the availability of the power plant has to fit with this demand. Lignite fired baseload plants usually fulfil this requirement.

7.2.4 Emergency situations

Considering the UPSWING concept, there is of course the risk of unplanned shutdown situations of either the waste incinerator or the connected power plant, e.g. caused by boiler trips. There are no major problems expected in case of a shutdown of the waste incinerator, as this situation is common practice in conventional MSWI. However, there will be a significant problem if the power plant must be shut down. In this case, the injection of waste flue gas has to be stopped immediately. A solution could be that the waste incinerator is coupled to more than one boiler, as already mentioned before. Furthermore, the waste incinerator should be equipped with an emergency chimney, which is also common practice in conventional MSWI, in order to cope with trips in the air pollution control system (APC). The control of the flue gas flow to the emergency chimney and to the coal boiler(s) can be achieved using a system of actuated controls and isolation dampers.

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[70] W.R. LIVINGSTON: "Start-up/shutdown, emergency operations and sensitivity/redundancy analysis", Project Deliverable D6, UPSWING, ENK5-CT-2002-00697, http://www.eu-projects.de/upswing

Regarding the SRF co-combustion concept, there are no potential problems expected in case of emergency situations. In case of boiler trips, the feeding of the additional fuel can be easily stopped. Considering problems in the SRF feeding system, the missing heat input can be compensated with an increased coal feed rate. In case of emergency situations, the co-combustion concept obviously has its advantages.

7.2.5 Environmental regulation

At the present time, it is considered that the coal-fired power plant involved in the UPSWING Process would have to comply with the requirements of the Waste Incineration Directive (Directive 2000/76/EC), rather than those of the Large Combustion Plant Directive (Directive 2001/80/EC), because of the general nature of the process and the introduction of waste flue gases containing PCDD/F to a coal fired boiler. Nevertheless, a final decision on this aspect strongly depends on the permitting authorities. Considering direct SRF co-combustion, the Waste Incineration Directive has to be applied anyway, regulating both mono-combustion of MSW in dedicated facilities and co-combustion of SRF in industrial furnaces or thermal power plants.

7.3 Economical assessment

The economical aspects have been highlighted in the introduction as all new and advanced waste treatment concepts - despite all potential environmental benefits - have to compete on the market. This question should be briefly discussed in the following section by means of the so-called “gate-fee”, the actual or estimated costs for waste disposal, usually expressed in €/Mg. Considering the UPSWING process, the necessary background was elaborated within the scope of the European Research Project UPSWING [71]. The information obtained for SRF production bases on direct contact to a German fuel producer, although detailed numbers and figures are not available.

7.3.1 Economical assessment of the UPSWING concept

To assess the economic benefits of the UPSWING concept the application was calculated for two different cases, namely a power station in Poland and in the Czech Republic. For both cases, the realisation of the UPSWING concept was compared with a conventional MSWI of the same size. The most important results of the cost comparison between the UPSWING process and a standalone MSWI process at the same location is the expected gate-fee for waste disposal in €/Mg. The results of this study [72] are summarised in Table 7-1 and Table 7-2:

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[71] UPSWING: “Unification of Power Plant and Solid Waste Incineration on the Grate”, ENK5-CT-2002-00697, http://www.eu-projects.de/upswing, (2003-2005)

[72] M. ANASTASIU, C. BOITA: “Results of costs comparison between coupled and stand-alone operations”, Project Deliverable D13, UPSWING, ENK5-CT-2002-00697, http://www.eu-projects.de/upswing

Table 7-1: Cost comparison standalone MSWI / UPSWING concept [72]

Case Study	Case : Poland I		Case : Poland II	
Waste throughput (Mg/a)	50,000		150,000	
Option	MSWI	UPSWING	MSWI	UPSWING
Electricity (€/kWh)	0.045		0.045	
Heat price (€/GJ)	4.2		4.2	
Coal price (€/GJ)	▨	15	▨	15
Annual benefits (Mio. €/a)	1.153	1.158	3.50	4.74
Gate-fee (€/Mg)	67.4	66.3	42.3	32.3
Cost reduction potential, %	0%	1.6%	0%	23.6%

Considering the potential of the UPSWING concept, the results for the investigated Polish power station show a significant dependency of the realisable gate-fee and the size of the connected waste incinerator. The benefit is almost zero for an annual capacity of 50,000 Mg (less than 2%), whereas for an annual capacity of 150,000 Mg a significant reduction seems to be possible (larger than 20%). An explanation for this difference is that steam and flue gas integration into an existing, already optimised power plant system resulted in decreases in the electrical efficiency compared to a stand-alone MSWI as determined by Ploumen [73]. Nevertheless, the annual cost and investment savings of the UPSWING process more than compensates for these losses. Hence, the larger the coupled waste incinerator, the larger the annual cost and investment savings, and the lower the negative effects in overall efficiency. However, there are technical limits regarding the amount of steam as well as flue gas integrated in the power plant process.

Considering the potential benefits of the concept in the Czech Republic, one can see a significant lower gate fee, even for a small-size waste incinerator:

Table 7-2: Cost comparison standalone MSWI / UPSWING concept [72]

Case Study	Case : Czech Republic	
Waste throughput (Mg/a)	50,000	
Option	MSWI	UPSWING
Electricity (€/kWh)	0.035	
Heat price (€/GJ)	4.1	
Coal price (€/GJ)	▨	10.2
Annual benefits (Mio. €/a)	1.24	1.96
Gate fee (€/Mg)	64.9	46.7
Cost reduction potential, %	0%	28.0%

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[73] P. PLOUMEN, R. SMEETS: “Efficiency improvement by integration of water/steam cycle of a MSWI with an existing power plant”, Project Deliverable D5, UPSWING, ENK5-CT-2002-00697, http://www.eu-projects.de/upswing

This result can be explained with lower electricity and heat revenues in combination with lower coal prices in the Czech Republic. As a consequence, the deficits in electrical efficiency are of lesser significance. Hence, the economic performance of the UPSWING process applied here would be even attractive for a small-scale waste incinerator.

7.3.2 Economical assessment of direct SRF co-combustion

In order to assess the economical perspective of direct SRF co-combustion, it is necessary to take a closer look on the fuel production process. The target of this production process is to generate a fuel from mixed waste materials, suitable for direct utilisation in the power plant process. The incoming waste is processed in various process steps, comprising of crushing, classifying, ferrous/non-ferrous separation, automatic sorting, and biological drying. High-calorific waste fractions (HCF) such as paper, cardboard, textiles, and plastics, are sorted out and further processed into a transportable (and combustible) form. Nevertheless, sorting means that not all the waste input material is suitable for the production of SRF, leaving different residual fractions which are to be treated correctly. To give a rough idea of the process, the following fractions can be expected during the production of SRF from MSW:

- High-calorific fraction suitable for SRF production: 30-35%
- Ferrous/non-ferrous material for recycling and losses: ~ 10%
- Water (evaporated during biological treatment): ~ 20%
- Residues (thermal treatment in MSWI): 35-40%

It should be clear that the fuel producer has to pay for the residual fraction to be disposed of in conventional MSWI. He will possibly get some revenues for ferrous/non-ferrous materials, but - most important - he cannot expect any significant revenues for the produced SRF. Taking this into account, a likely question will be if the production of SRF is economical sound at all. Indeed it is, which becomes clear when considering the reduction of the initial waste input mass: Only 35-40% have to be treated in conventional MSWI, which means a significant reduction of 60-65% compared to the initial mass. Nevertheless, the fuel producer gets paid for collection and treatment of the entire waste input. Accordingly, his total calculation depends on the quota of SRF produced and its quality, and strictly connected, upon the reduction of the residual fraction to be disposed off in conventional MSWI. To give a rough estimation on the required production costs, it is likely that the fuel producer has to pay between 90 and 110 €/Mg (based on a private communication with a German SRF producer), depending on the quality of the waste input and the subsequent SRF quota of his SRF production plant.

7.3.3 Comparison of the investigated concepts

For the UPSWING process, an economical benefit in comparison to conventional MSWI was shown based on two dedicated case studies elaborated within the scope of the European Research Project UPSWING. This benefit will be strictly connected to the incinerator size and the annual cost and investment savings in comparison to a conventional MSWI. As deficits in the overall efficiency of the power plant are expected, electricity and heat revenues have to be taken into account as well. Nevertheless, the annual cost and investment savings of the UPSWING process more than compensates for these losses.

Considering the SRF production process, a direct comparison of a potential economical benefit in comparison to conventional MSWI is rather difficult. There are different fuel production technologies available on the market and an assessment of the expected costs for SRF production is not part of the present thesis. Considering their utilisation in the power plant, a considerable benefit can be determined from the view of the power plant operator. These benefits derive from the production of electricity or heat from SRF and by the substitution of primary fuel, and possibly from CO_2 savings if the biodegradable share of SRF is considered CO_2-neutral. Again, exact figures of the possible savings cannot be given as a cost-benefit analysis of SRF combustion is not part of this thesis.

7.4 Conclusions and future outlook

In this thesis, the environmental, operational and economic performance of the UPSWING concept was evaluated by means of a process comparison with other state-of-the-art concepts, viz. conventional municipal solid waste incineration and direct co-combustion of waste-derived fuels in thermal power plants. Environmental and operational aspects were investigated by experimental and theoretical means. Furthermore, the economical aspects were theoretically assessed by a comparison of the expected costs for the investigated concepts. Following this approach, it was possible to identify benefits respectively deficits of the different waste disposal concepts, and to identify crucial aspects and to assess their relevance.

Considering environmental aspects, it was found that the UPSWING process is generally capable to achieve and to maintain the high standards of conventional municipal solid waste incineration. This assessment bases on the results acquired in chapter 5, showing a good control of waste-specific pollutants and their abatement in the partial flue gas cleaning system. In view of the pollutants NO_x, SO_2, and PCDD/F, which are not removed prior to waste flue gas injection, it was found that they can be safely reduced or destroyed in the power plant process. Regarding direct SRF co-combustion, a certain amount of pollutants will be brought into the combustion process. Heavy metals should be mentioned in this context, although the concentration found in the SRF strongly depend on the origin of the input materials and the quality of the fuel production process itself. Their abatement in the process, for example via the fly ashes or the FGD system, should be ensured as well. Nevertheless, a dilution of the residues from coal combustion with waste-borne pollutants has to be accepted. From an environmental point of view, especially in terms of reliable pollutant control, the UPSWING process definitely has its advantages.

Considering the operational performance of both processes, especially in terms of process integration (waste flue gas injection vs. SRF handling and feeding), heat recovery, flexibility of plant operation, and emergency situations, a clear advantage of the co-combustion concept was found. Generally, the operation of the integrated UPSWING process appears to be more difficult. Two plants have to be operated simultaneously, and this will reduce the flexibility of plant operation in any case. Operational problems such as boiler trips or problems with the air pollution control system will always have severe consequences on one or the other plant. Furthermore, the efforts for control and regulation will be significantly higher in comparison to standalone processes. Considering heat recovery, the additional waste flue gases will most likely influence the overall process efficiency. Contrary to this, the application of direct SRF co-combustion implies much lesser efforts in terms of plant operation. Using a separate dosing and feeding system, with possible injection via separate burners, allows a controlled injection of SRF during normal plant operation. Favourable is the injection of a fixed amount, correlated to the boiler load rate, and to rely on the standard load control via the coal feeding rate. In case of critical operational situations such as boiler start-up or shutdown phases, the injection of the secondary fuel can be stopped. Hence, the co-combustion concept proposes a far better freedom of boiler operation.

Regarding the economical performance, the main benefit of the UPSWING process is the incineration of untreated MSWI in a conventional grate firing system. This technology is common practice and different suppliers are available on the market. As a consequence of flue gas and steam integration, certain components in the flue gas treatment system as well as the generator unit can be omitted, which results in general lower investment costs and taking the cost for integration into account. In terms of SRF co-combustion, the secondary fuel has to be produced and transported to the power plant location. At the power plant site itself, additional investment costs are necessary for storage and feeding installations as well as for distribution and injection of the material into the furnace. From the view of the power plant operation, the additional investment costs are considerably low. The main costs for the entire fuel production process will be covered by the fuel producer.

The final question would be if the increased efforts for the UPSWING concept are worth to be taken into consideration. To conclude with a personal view, this answer would be yes, especially from an environmental point of view. Accordingly, a large-scale demonstration of the UPSWING process would be the next step to evaluate its capability, especially in terms of its environmental, operational, and economical performance.

Appendix A-1

a) Proximate and ultimate analysis of the investigated coals

Table A-1/1: Proximate and ultimate fuel analysis

Parameter	Unit	B/1	B/2	B/3	L/1	L/2	L/3
Characterisation	-	Bituminous coal	Bituminous coal	Bituminous coal	Lignite	Hard brown coal	Brown coal
Water	%	2.4	1.2	1.0	4.2	4.2	4.9
Ash	$\%_{\mathrm{dry}}$	15.5	8.3	5.4	35.5	19.7	10.9
Volatiles	$\%_{\mathrm{daf}}$	40.7	37.1	32.2	60.3	54.0	58.1
Fixed Carbon	$\%_{\mathrm{daf}}$	59.3	62.9	67.8	39.7	46.0	41.9
Carbon	$\%_{\mathrm{daf}}$	79.5	84.8	86.2	65.9	74.8	65.5
Hydrogen	$\%_{\mathrm{daf}}$	6.1	6.3	6.0	7.3	5.7	6.0
Sulphur	$\%_{\mathrm{daf}}$	0.6	0.7	0.7	1.0	0.9	0.4
Chlorine	$\%_{\mathrm{daf}}$	0.05	0.36	0.19	0.06	0.06	0.05
Nitrogen	$\%_{\mathrm{daf}}$	1.5	1.7	1.5	0.6	0.9	0.6
Oxygen *	$\%_{\mathrm{daf}}$	12.4	6.2	5.4	25.2	17.7	27.5
NCV	MJ/kg	25.965	30.836	32.438	15.689	19.468	21.146

* oxygen calculated   dry: water free   daf: dry- and ash free

b) Major and minor ash elements of the investigated coals

Table A-1/2: Major and minor ash elements

Element	Unit	B/1	B/2	B/3	L/1	L/2	L/3
Characterisation	-	Bituminous coal	Bituminous coal	Bituminous coal	Lignite	Hard brown coal	Brown coal
SiO_2	$\%_{ash}$	59.7	35.5	45.2	48.3	45.0	49.2
Al_2O_3	$\%_{ash}$	20.3	20.2	29.5	31.2	29.9	5.0
CaO	$\%_{ash}$	1.9	10.5	3.4	1.4	2.6	20.0
MgO	$\%_{ash}$	2.0	2.7	1.9	1.0	0.9	3.8
K_2O	$\%_{ash}$	2.3	1.8	3.5	1.6	0.5	0.3
Na_2O	$\%_{ash}$	1.0	1.0	1.2	1.3	1.3	0.3
Fe_2O_3	$\%_{ash}$	7.0	16.2	8.3	5.6	8.1	12.0
P_2O_5	$\%_{ash}$	0.2	0.1	0.9	0.1	0.4	0.1
TiO_2	$\%_{ash}$	0.9	0.7	1.1	1.3	6.0	1.5
MnO	$\%_{ash}$	< 0.1	0.2	< 0.1	< 0.1	< 0.1	0.2
SO_3^*	$\%_{ash}$	1.3	10.5	2.5	1.6	1.7	7.3

* calculated from sulphur acquired from elemental ash analysis

c) Trace element concentrations in the investigated coals

Table A-1/3: Trace element concentration

Element	Unit	B/1	B/2	B/3	L/1	L/2	L/3
Characterisation	-	Bituminous coal	Bituminous coal	Bituminous coal	Lignite	Hard brown coal	Brown coal
Hg	mg/kg_{coal}	0.12	0.24	0.25	0.40	0.63	0.14
As	mg/kg_{coal}	3.4	▨	▨	< 2.7	14.0	< 1.0
Pb	mg/kg_{coal}	2.9	11.0	9.0	10.6	4.8	< 1.5
Cd	mg/kg_{coal}	0.26	0.20	0.10	< 0.15	0.15	< 0.15
Cr	mg/kg_{coal}	22.0	▨	▨	36.0	39.0	< 10.0
Cu	mg/kg_{coal}	7.7	7.0	13.0	11.4	75.0	3.4
Ni	mg/kg_{coal}	12.0	17.0	20.0	18.5	14.0	< 3.0
Zn	mg/kg_{coal}	93.0	15.0	2.0	29.8	55.0	3.9

Appendix A-2

a) Proximate and ultimate analysis of the investigated SRF

Table A-2/1: Proximate and ultimate analysis

Parameter	Unit	SRF/04	SRF/05	SRF/PRR
Water	%	2.6	2.5	3.2
Ash	$\%_{dry}$	12.9	11.5	8.3
Volatiles	$\%_{daf}$	90.4	87.7	92.8
Fixed Carbon	$\%_{daf}$	9.6	12.3	7.2
Carbon	$\%_{daf}$	58.6	54.5	62.2
Hydrogen	$\%_{daf}$	9.6	8.4	9.2
Sulphur	$\%_{daf}$	0.1	0.2	0.05
Chlorine	$\%_{daf}$	0.34	0.64	1.13
Nitrogen	$\%_{daf}$	0.78	0.95	0.2
Oxygen *	$\%_{daf}$	30.6	35.5	27.2
NCV	kJ/kg	22850	20770	22800
* oxygen calculated dry: water free daf: dry- and ash free				

b) Major and minor ash elements of the investigated SRF

Table A-2/2: Major and minor ash elements

Parameter	Unit	SRF/04	SRF/05	SRF/PRR
SiO_2	$\%_{ash}$	23.0	31.9	30.3
Al_2O_3	$\%_{ash}$	45.9	17.3	19.8
CaO	$\%_{ash}$	20.8	31.0	24.4
MgO	$\%_{ash}$	1.5	2.7	2.3
K_2O	$\%_{ash}$	1.0	2.0	0.6
Na_2O	$\%_{ash}$	2.7	4.4	2.3
Fe_2O_3	$\%_{ash}$	1.2	2.6	13.6
P_2O_5	$\%_{ash}$	1.2	1.4	0.4
TiO_2	$\%_{ash}$	1.6	2.7	4.5
MnO	$\%_{ash}$	< 0.1	< 0.1	0.2
SO_3	$\%_{ash}$	0.9	2.7	2.6

c) Trace element concentrations in the investigated SRF

Table A-2/3: Trace elements

Element	Unit	SRF/04	SRF/05	SRF/PRR
Hg	mg/kg$_{ds}$	0.10	0.16	0.20
As	mg/kg$_{ds}$	< 0.7	0.88	▨
Pb	mg/kg$_{ds}$	24.3	58.3	56.0
Cd	mg/kg$_{ds}$	1.36	0.96	2.5
Cr	mg/kg$_{ds}$	27.1	73.6	▨
Cu	mg/kg$_{ds}$	35.1	639.9	280.0
Ni	mg/kg$_{ds}$	9.22	43.6	84.0
Zn	mg/kg$_{ds}$	120.0	308.6	245.0

Appendix B-0 : Basic input/output balances

a) General process balances

A mass balance was formulated covering all relevant input streams into the combustion process. This approach allows the quantification of specific elements such as chlorine, sulphur, or mercury in the process output streams, comprising of flue gas and fly ash. Fig. B-0/1 shows the initial input- and output streams as well as the respective concentrations:

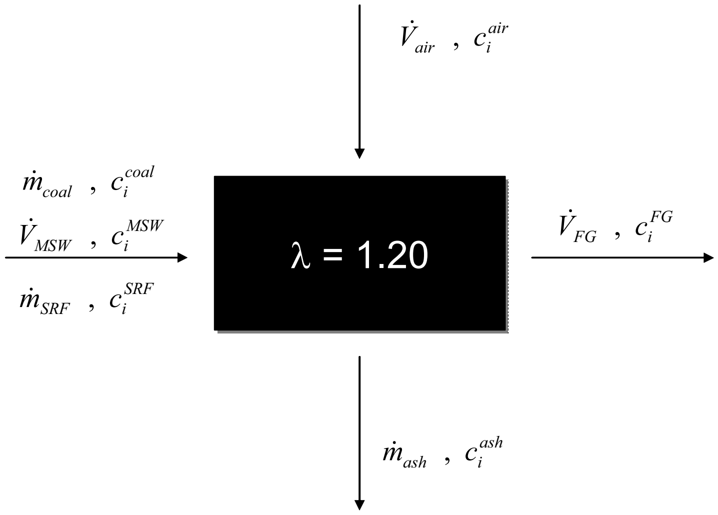


Fig. B-0/1: Basic input- / output balance

The excess air ratio of λ = 1.20 at the reactor exit can be considered as a fixed parameter as it was maintained during all experimental trials. Input streams include coal as the primary fuel, combustion air, and - in addition - waste flue gases or co-fired SRF material. Process output streams include flue gases and combustion ashes. In case of SRF co-combustion, the residual combustion ash will be a mixed ash from both input fuels. In case of the UPSWING process, the waste flue gases contain no dust or particulate matter as a result of a complete ash removal in the partial flue gas cleaning system. Following these considerations the **mass input** of an element i can be calculated according to equation B.1:

$$\dot{m}_i^{input} = c_i^{coal} \cdot \dot{m}_{coal} + c_i^{SRF} \cdot \dot{m}_{SRF} + c_i^{MSWI} \cdot \dot{V}_{MSWI} + c_i^{air} \cdot \dot{V}_{air} \qquad \text{mg/h} \qquad (B.1)$$

As a first estimation of an element concentration in the ash, a **maximum ash concentration $c_{i,max}^{ash}$** (mg/kg) of an element i can be calculated according to equation B.2:

$$c_{i,\max}^{ash} = \frac{\dot{m}_i^{input}}{\dot{m}_{ash}} \qquad \text{mg/kg} \qquad (B.2)$$

In order to evaluate the fraction of an element i bound to fly ash particles, the **recovery rate $\mathbf{R_i^{ash}}$** (%) can be calculated as the ratio of input and output:

$$R_i^{ash} = \frac{c_i^{ash} \cdot \dot{m}_{ash}}{\dot{m}_i^{input}} = \frac{c_i^{ash} \cdot (\dot{m}_{coal} \cdot c_{ash}^{coal} + \dot{m}_{SRF} \cdot c_{ash}^{SRF})}{\dot{m}_i^{input}} \qquad \% \qquad \text{(B.3)}$$

The recovery rate is a useful indicator to evaluate the behaviour of an element in the combustion process. Due to the fact that the exact amount of ash separated in the ash filter system cannot be determined, the recovery rate was calculated on basis of the total ash input, which is calculated from the ash content of the combusted fuel(s). This approach may lead to recovery rates larger than 100%. Correspondent results are not necessarily wrong and can be explained with an enrichment of certain elements (heavy metals) in the ash filter system. Enrichment is usually caused by condensation of volatile elements on fine fly ash particles. The condensable part of a heavy metal is a function of its partial pressure and the temperature related vapour pressure of the element. Therefore, the temperature is a crucial factor in terms of condensation, as well as other factors such as particle size, deposition rate and diffusion processes.

Following the considerations for a recovery rate of an element i in the fly ash, a comparable approach can be done for an element in the flue gas, although an enrichment of an element cannot be expected. A (theoretical) **maximum flue gas concentration $\mathbf{c_{i,max}^{FG}}$** (mg/m³) of an element i can be calculated according to equation B.4:

$$c_{i,\max}^{FG} = \frac{\dot{m}_i^{input}}{\dot{V}_{FG}} \qquad \text{mg/m}^3 \qquad \text{(B.4)}$$

Based on the measured flue gas concentration c_i^{FG}, a **recovery rate $\mathbf{R_i^{FG}}$** (%) can be calculated for an element i in the flue gas:

$$R_i^{FG} = \frac{c_i^{FG}}{c_{i,\max}^{FG}} = \frac{c_i^{FG} \cdot \dot{V}_{FG}}{\dot{m}_i^{input}} \qquad \% \qquad \text{(B.5)}$$

b) Balancing the high-volatile element mercury

To distinguish between the different flue gas species, it is essential to assess its behaviour and environmental potential of mercury in the process. Both gaseous $HgCl_2(g)$ and total gaseous mercury Hg(g) were measured during trials at the test facilities. Assuming gaseous elemental $Hg^0(g)$ and gaseous $HgCl_2(g)$ as the primary mercury species, the concentration of gaseous elemental $Hg^0(g)$ can be calculated as the difference between total gaseous mercury Hg(g) and gaseous $HgCl_2(g)$:

$$c^{FG}_{Hg^0(g)} = c^{FG}_{Hg(g)} - c^{FG}_{HgCl_2(g)} \qquad \mu g/m^3 \qquad (B.6)$$

To evaluate the quality of the measurement, a **recovery rate R_{Hg}** (%) can be calculated considering total gaseous and particle-bound mercury in relation to the total mercury input:

$$R_{Hg} = \frac{c^{FG}_{Hg(g)} \cdot \dot{V}_{FG} + c^{ash}_{Hg} \cdot \dot{m}_{ash}}{\dot{m}^{input}_{Hg}} \qquad \% \qquad (B.7)$$

In this context it is of interest to evaluate the mass fractions of the different species, which can calculated according to equations B.8 – B.10:

$$Hg^0(g) = \frac{c^{FG}_{Hg^0(g)} \cdot \dot{V}_{FG}}{\dot{m}^{input}_{Hg}} \qquad \% \qquad (B.8)$$

$$HgCl_2(g) = \frac{c^{FG}_{HgCl_2(g)} \cdot \dot{V}_{FG}}{\dot{m}^{input}_{Hg}} \qquad \% \qquad (B.9)$$

$$Hg(p) = \frac{c^{ash}_{Hg} \cdot \dot{m}_{ash}}{\dot{m}^{input}_{Hg}} \qquad \% \qquad (B.10)$$

During several trials performed, it was found that the recovery rates differed significantly with the consequence that a direct comparison of the results is not possible. A normalisation of the individual results appears to be suitable to solve this deficit. The normalised concentration can be calculated according to B.11:

$$\frac{Hg^0(g) + HgCl_2(g) + Hg(p)}{R^{total}_{Hg}} = 100\% \qquad (B.11)$$

Appendix B-1 : Basic balances for the UPSWING

UPSWING flue gas balances

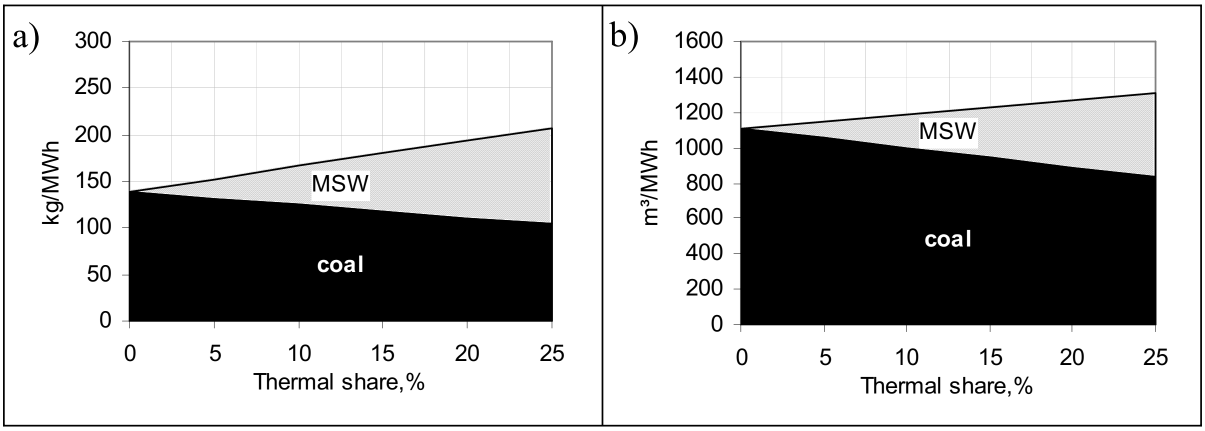


Fig. B-1/1: Bituminous coal B.1 - MSW a) mass throughput kg/MWh b) flue gas volume m³/MWh

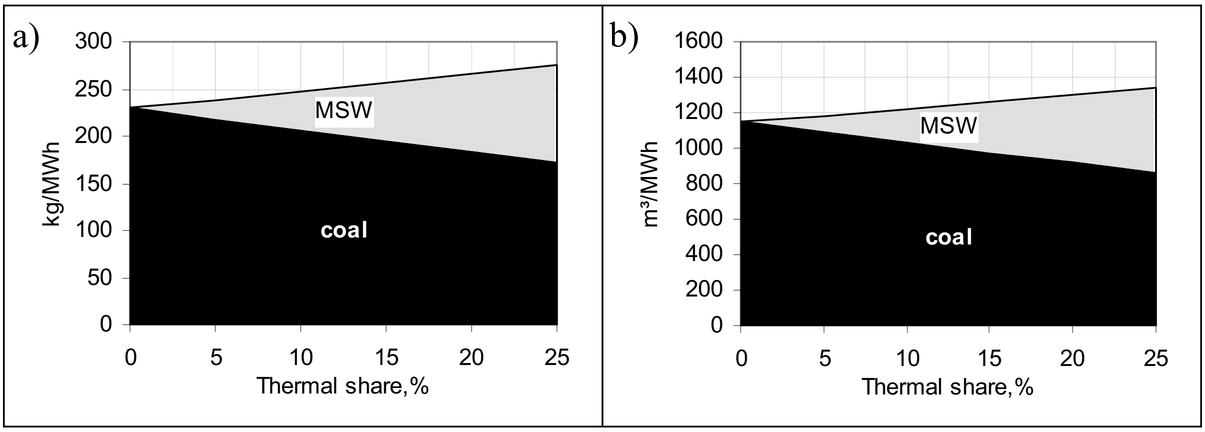


Fig. B-1/2: Lignite L.1 - MSW a) mass throughput kg/MWh b) flue gas volume m³/MWh

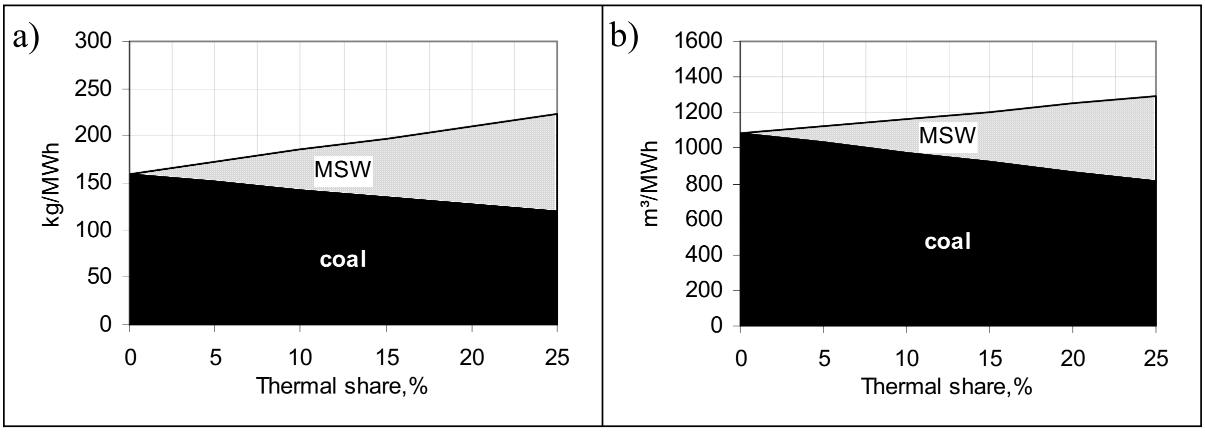


Fig. B-1/3: Lignite L.2 - MSW a) mass throughput kg/MWh b) flue gas volume m³/MWh

Appendix B-2 : Basic balances for SRF co-combustion

I. Basic input balances

Table B-2/1: Mass- and input balance bituminous coal B.1 - SRF/04

SRF/04.B1	m_{coal}, kg/h	m_{SRF}, kg/h	V_{FG}, m³/h	m_{ash}, g/h	m^{in}_{S}, g/h	m^{in}_{Cl}, mg/h	m^{in}_{Hg}, µg/h
0% SRF	0.800	0.000	6.40	121.4	4.00	320.00	96.00
5% SRF	0.768	0.042	6.42	121.8	3.87	427.47	96.36
10% SRF	0.736	0.085	6.45	122.3	3.74	537.80	96.82

Table B-2/2: Mass- and input balance brown coal L.3 - SRF/04

SRF/04.L3	m_{coal}, kg/h	m_{SRF}, kg/h	V_{FG}, m³/h	m_{ash}, g/h	m^{in}_{S}, g/h	m^{in}_{Cl}, mg/h	m^{in}_{Hg}, µg/h
0% SRF	1.200	0.000	7.80	124.8	3.60	480.00	168.00
5% SRF	1.152	0.043	7.77	125.2	3.49	583.93	165.58
10% SRF	1.104	0.086	7.74	125.6	3.37	687.87	163.16

Table B-2/3: Mass- and input balance brown coal L.3 - SRF/05

SRF/05.L3	m_{coal}, kg/h	m_{SRF}, kg/h	V_{FG}, m³/h	m_{ash}, g/h	m^{in}_{S}, g/h	m^{in}_{Cl}, mg/h	m^{in}_{Hg}, µg/h
0% SRF	1.200	0.000	7.80	124.8	3.60	480.00	168.00
5% SRF	1.152	0.043	7.74	124.6	3.53	699.77	168.16
10% SRF	1.104	0.086	7.68	124.5	3.46	919.55	168.32

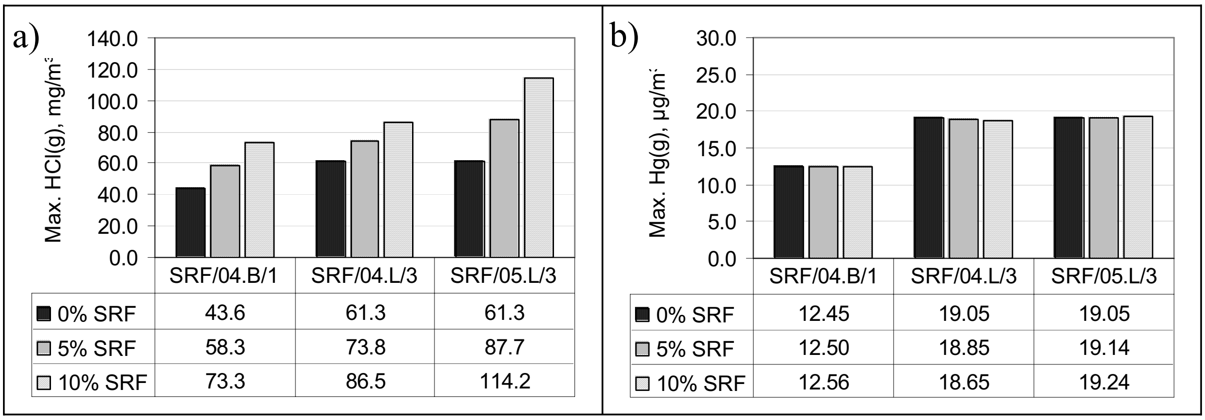
Table B-2/4: Mass- and input balance bituminous coal B.2 - SRF/PRR

SRF/PRR.B2	m_{coal}, kg/h	m_{SRF}, kg/h	V_{FG}, m³/h	m_{ash}, g/h	m^{in}_{S}, g/h	m^{in}_{Cl}, mg/h	m^{in}_{Hg}, µg/h
0% SRF	35.000	0.000	332.50	2882.3	218.93	115500.0	8400.0
5% SRF	33.250	2.370	333.18	2928.8	208.93	133424.1	8454.0
10% SRF	31.500	4.750	333.93	2976.2	198.93	151448.3	8510.0

Table B-1/5: Mass- and input balance bituminous coal B.3 - SRF/PRR

SRF/PRR.B3	m_{coal}, kg/h	m_{SRF}, kg/h	V_{FG}, m³/h	m_{ash}, g/h	m^{in}_{S}, g/h	m^{in}_{Cl}, mg/h	m^{in}_{Hg}, µg/h
0% SRF	33.500	0.000	331.65	1804.0	230.15	60300.0	8375.0
5% SRF	31.820	2.370	332.32	1904.2	219.55	80975.1	8429.0
10% SRF	30.150	4.750	333.16	2005.8	209.03	101768.3	8487.5

II. Balanced maximum flue gas concentrations



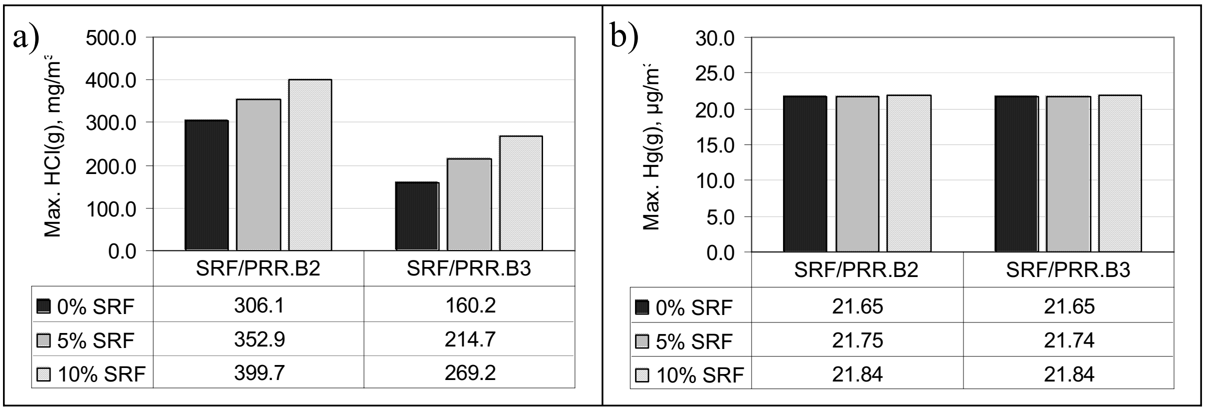
a)

	SRF/04.B/1	SRF/04.L/3	SRF/05.L/3
■ 0% SRF	43.6	61.3	61.3
▨ 5% SRF	58.3	73.8	87.7
□ 10% SRF	73.3	86.5	114.2

b)

	SRF/04.B/1	SRF/04.L/3	SRF/05.L/3
■ 0% SRF	12.45	19.05	19.05
▨ 5% SRF	12.50	18.85	19.14
□ 10% SRF	12.56	18.65	19.24

Fig. B-1/1: Balanced maximum concentration for SRF/04 and SRF/05 a) HCl(g) b) Hg(g)



a)

	SRF/PRR.B2	SRF/PRR.B3
■ 0% SRF	306.1	160.2
▨ 5% SRF	352.9	214.7
□ 10% SRF	399.7	269.2

b)

	SRF/PRR.B2	SRF/PRR.B3
■ 0% SRF	21.65	21.65
▨ 5% SRF	21.75	21.74
□ 10% SRF	21.84	21.84

Fig. B-1/2: Balanced maximum concentration for SRF/PRR a) HCl(g) b) Hg(g)

Appendix C-1 : Trace elements UPSWING process

1. Combustion settings

Table C-1/1: Basic combustion settings for the investigated coals

Parameter / Coal	Coal B/1	Coal L/1	Coal L.2
Coal mass flow	1.00 kg/h	1.00 kg/h	1.00 kg/h
Reactor temperature	1100°C	1100°C	1100°C
Combustion air (λ=1.20)	8.46 m³/h	5.37 m³/h	7.15 m³/h
Flue gas coal (dry)	8.08 m³/h	5.16 m³/h	6.86 m³/h
Flue gas coal (wet)	8.86 m³/h	5.71 m³/h	7.54 m³/h
10% MSWI flue gas (dry)	0.81 m³/h	0.52 m³/h	0.69 m³/h
Additional NO load	0, 200, 400 ppm	0, 200, 400 ppm	0, 200, 400 ppm
Additional H_2O load	0.38 kg/m³	0.38 kg/m³	0.38 kg/m³

Table C-1/2: Combustion air and flue gas amount for additional flue gas injection (Case A)

Case A (additional)	Coal B/1	Coal L/1	Coal L/2
Combustion air	8.46 m³/h	5.37 m³N/h	7.15 m³/h
Flue gas total (dry)	8.89 m³/h	5.67 m³N/h	7.55 m³/h
Flue gas total (wet)	9.86 m³/h	6.34 m³N/h	8.39 m³/h
O2 level at furnace exit	4.1 vol.-%	4.1 vol.-%	4.1 vol.-%

Table C-1/3: Combustion air and flue gas amount for substitution of combustion air (Case B)

Case B (substitution)	Coal B/1	Coal L/1	Coal L/2
Combustion air	8.06 m³/h	5.17 m³N/h	6.81 m³/h
Substituted combustion air	0.40 m³/h	0.20 m³N/h	0.34 m³/h
Flue gas total (dry)	8.49 m³/h	5.56 m³N/h	7.22 m³/h
Flue gas total (wet)	9.46 m³/h	6.24 m³N/h	8.05 m³/h
O2 level at furnace exit	3.5 vol.-%	3.5 vol.-%	3.5 vol.-%

2. Nomenclature

2.1 Trials with the Polish lignite L/1

Table C-1/4: Lignite L/1, 1100°C

λ ↓ / Hg →	Baseline dry	0 μg/m^3_N	100 μg/ m^3_N	200 μg/ m^3_N
1.20	V-001 [A]	V-111 [A]	V-112	V-113 [A]
0.95	V-002	V-121	V-122	V-123
0.85	V-003 [A]	V-131 [A]	V-132	V-133 [A]

[A] Fly ash sampled and analysed

Table C-1/5: Lignite L/1, 1300°C

λ ↓ / Hg →	Baseline dry	0 μg/m^3_N	100 μg/ m^3_N	200 μg/ m^3_N
1.20	V-311 [A]	V-312 [A]	V-313	V-314 [A]
0.95	V-321	V-322	V-323	V-324
0.85	V-331 [A]	V-332 [A]	V-333	V-334 [A]

[A] Fly ash sampled and analysed

Table C-1/6: Lignite L/1, 1100°C

HCl ↓ / Hg →	0 μg/m^3_N	100 μg/ m^3_N	200 μg/ m^3_N
0 mg/m^3_N	V-211 / Hg-111 [A]	V-212 / Hg-112	V-213 / V-113 [A]
400 mg/m^3_N	V-221 [A]	V-222	V-223 [A]
800 mg/m^3_N	V-231 [A]	V-232	V-233 [A]

[A] Fly ash sampled and analysed

Table C-1/7: Lignite L/1, 1100°C

λ ↓ / Hg →	Baseline dry	0 μg/m^3_N	100 μg/ m^3_N	200 μg/ m^3_N
1.20	V-511 [A]	V-512 [A]	V-513	V-514 [A]
0.95	V-521	V-522	V-523	V-524
0.85	V-531 [A]	V-532	V-533 [A]	V-534 [A]

[A] Fly ash sampled and analysed

Table C-1/8: Lignite L/1, 1300°C

λ ↓ / Hg →	Baseline dry	0 μg/m^3_N	100 μg/ m^3_N	200 μg/ m^3_N
1.20	V-411 [A]	V-412 [A]	V-413	V-414 [A]
0.95	V-421	V-422	V-423	V-424
0.85	V-431 [A]	V-432 [A]	V-433	V-434 [A]

[A] Fly ash sampled and analysed

2.2 Trials with the Colombian bituminous coal B/1

Table C-1/9: Bituminous coal B/1, 1100°C

λ ↓ / Hg →	Baseline dry	0 µg/m^3_N	100 µg/ m^3_N	200 µg/ m^3_N
1.20	V-511 [A]	V-512 [A]	V-513	V-514 [A]
0.95	V-521	V-522	V-523	V-524
0.85	V-531 [A]	V-532	V-533 [A]	V-534 [A]
[A] Fly ash sampled and analysed				

Table C-1/10: Bituminous coal B/1, 1300°C

λ ↓ / Hg →	Baseline dry	0 µg/m^3_N	100 µg/ m^3_N	200 µg/ m^3_N
1.20	V-411 [A]	V-412 [A]	V-413	V-414 [A]
0.95	V-421	V-422	V-423	V-424
0.85	V-431 [A]	V-432 [A]	V-433	V-434 [A]
[A] Fly ash sampled and analysed				

3. Results for gaseous and particle-bound mercury

3.1 Results of the trials with the Polish lignite L/1

Table C-1/11: Lignite L/1, 1100°C, λ = 1.20

V	c_{HgCl2}^{FG}	c_{Hg0}^{FG}	c_{Hg}^{FG}	c_{Hg}^{ash}	R_{Hg}^{tot}	$HgCl_2(g)$	$Hg^0(g)$	Hg(p)
-	µg/m³	µg/m³	µg/m³	µg/kg	%	%	%	%
001	17.5	39.5	57.0	230.0	90.8%	24.1%	54.4%	21.5%
111	9.0	46.0	55.0	150.0	95.3%	14.2%	72.4%	13.4%
113	20.5	64.5	85.0	140.0	92.9%	22.1%	69.4%	8.5%

Table C-1/12: Lignite L/1, 1100°C, λ = 0.85

V	c_{HgCl2}^{FG}	c_{Hg0}^{FG}	c_{Hg}^{FG}	c_{Hg}^{ash}	R_{Hg}^{tot}	$HgCl_2(g)$	$Hg^0(g)$	Hg(p)
-	µg/m³	µg/m³	µg/m³	µg/kg	%	%	%	%
003	16.5	38.0	54.5	180.0	83.4%	24.7%	56.9%	18.3%
131	14.5	40.5	55.0	50.0	86.8%	25.1%	70.0%	4.9%
133	24.5	55.5	80.0	260.0	94.7%	25.9%	58.6%	15.6%

Table C-1/13: Lignite L/1, 1300°C, λ = 1.20

V	c_{HgCl2}^{FG}	c_{Hg0}^{FG}	c_{Hg}^{FG}	c_{Hg}^{ash}	R_{Hg}^{tot}	$HgCl_2(g)$	$Hg^0(g)$	Hg(p)
-	µg/m³	µg/m³	µg/m³	µg/kg	%	%	%	%
311	15.0	20.0	35.0	100.0	52.3%	35.9%	47.8%	16.3%
312	12.5	26.0	38.5	100.0	66.3%	28.3%	58.9%	12.8%
314	23.5	30.5	54.0	100.0	59.7%	39.4%	51.1%	9.5%

Table C-1/14: Lignite L/1, 1300°C, λ = 0.85

V	c_{HgCl2}^{FG}	c_{Hg0}^{FG}	c_{Hg}^{FG}	c_{Hg}^{ash}	R_{Hg}^{tot}	$HgCl_2(g)$	$Hg^0(g)$	Hg(p)
-	µg/m³	µg/m³	µg/m³	µg/kg	%	%	%	%
331	15.5	17.5	33.0	240.0	61.7%	31.4%	35.5%	33.1%
332	10.5	23.0	33.5	100.0	58.8%	26.8%	58.7%	14.5%
334	19.0	33.5	52.5	160.0	61.6%	30.9%	54.4%	14.7%

Table C-1/15: Lignite L/1, 1100°C, Hg = 0 µg/m³, HCl = 0, 400, 800 mg/m³

V	c_{HgCl2}^{FG}	c_{Hg0}^{FG}	c_{Hg}^{FG}	c_{Hg}^{ash}	R_{Hg}^{tot}	$HgCl_2(g)$	$Hg^0(g)$	Hg(p)
-	µg/m³	µg/m³	µg/m³	µg/kg	%	%	%	%
111	9.0	46.0	55.0	150.0	95.3%	14.2%	72.4%	13.4%
221	13.5	24.5	38.0	260.0	79.1%	25.6%	46.5%	27.9%
231	12.5	26.0	38.5	100.0	66.3%	28.3%	58.9%	12.8%

Table C-1/16: Lignite L/1, 1100°C, Hg = 200 µg/m³, HCl = 0, 400, 800 mg/m³

V	c_{HgCl2}^{FG}	c_{Hg0}^{FG}	c_{Hg}^{FG}	c_{Hg}^{ash}	R_{Hg}^{tot}	$HgCl_2(g)$	$Hg^0(g)$	Hg(p)
-	µg/m³	µg/m³	µg/m³	µg/kg	%	%	%	%
113	20.5	64.5	85.0	140.0	92.9%	22.1%	69.4%	8.5%
223	18.5	41.0	59.5	320.0	77.6%	23.8%	52.8%	23.4%
233	21.5	31.0	52.5	170.0	62.1%	34.6%	49.9%	15.5%

3.2 Results of the trials with the Colombian bituminous coal B/1

Table C-1/17: Colombian bituminous coal B/1, 1100°C, $\lambda = 1.20$

V	c_{HgCl2}^{FG}	c_{Hg0}^{FG}	c_{Hg}^{FG}	c_{Hg}^{ash}	R_{Hg}^{tot}	$HgCl_2$(g)	Hg^0(g)	Hg(p)
-	µg/m³	µg/m³	µg/m³	µg/kg	%	%	%	%
511	2.5	4.0	6.5	180.0	66.1%	25.2%	40.4%	34.4%
512	3.0	2.5	5.5	300.0	80.1%	28.7%	23.9%	47.4%
514	7.5	9.5	17.0	580.0	67.9%	28.2%	35.8%	36.0%

Table C-1/18: Colombian bituminous coal B/1, 1100°C, $\lambda = 0.85$

V	c_{HgCl2}^{FG}	c_{Hg0}^{FG}	c_{Hg}^{FG}	c_{Hg}^{ash}	R_{Hg}^{tot}	$HgCl_2$(g)	Hg^0(g)	Hg(p)
-	µg/m³	µg/m³	µg/m³	µg/kg	%	%	%	%
531	3.0	4.0	7.0	250.0	78.3%	25.6%	34.1%	40.4%
533	9.0	6.0	15.0	100.0	63.8%	54.1%	36.0%	9.9%
534	15.0	8.0	23.0	360.0	73.9%	51.8%	27.6%	20.5%

Table C-1/19: Colombian bituminous coal B/1, 1300°C, $\lambda = 1.20$

V	c_{HgCl2}^{FG}	c_{Hg0}^{FG}	c_{Hg}^{FG}	c_{Hg}^{ash}	R_{Hg}^{tot}	$HgCl_2$(g)	Hg^0(g)	Hg(p)
-	µg/m³	µg/m³	µg/m³	µg/kg	%	%	%	%
411	2.0	3.0	5.0	100.0	46.0%	29.0%	43.5%	27.5%
412	2.5	4.5	7.0	290.0	90.3%	21.2%	38.2%	40.6%
414	7.5	10.0	17.5	100.0	48.9%	39.2%	52.2%	8.6%

Table C-1/20: Colombian bituminous coal B/1, 1300°C, $\lambda = 0.85$

V	c_{HgCl2}^{FG}	c_{Hg0}^{FG}	c_{Hg}^{FG}	c_{Hg}^{ash}	R_{Hg}^{tot}	$HgCl_2$(g)	Hg^0(g)	Hg(p)
-	µg/m³	µg/m³	µg/m³	µg/kg	%	%	%	%
431	2.5	2.5	5.0	100.0	46.0%	36.3%	36.3%	27.5%
432	2.0	3.5	5.5	100.0	54.8%	28.0%	49.0%	23.1%
434	8.0	10.5	18.5	420.0	65.0%	31.5%	41.3%	27.2%

3.3 Influence of operational conditions on mercury speciation (lignite L/1)

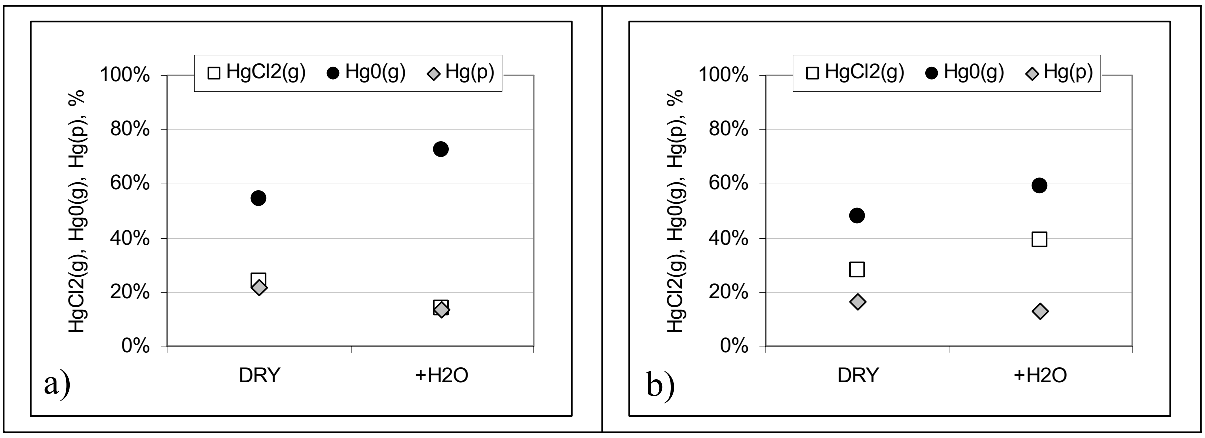


Fig. C-1/1: Influence of additional water on mercury speciation (unstaged) a) 1100°C b) 1300°C

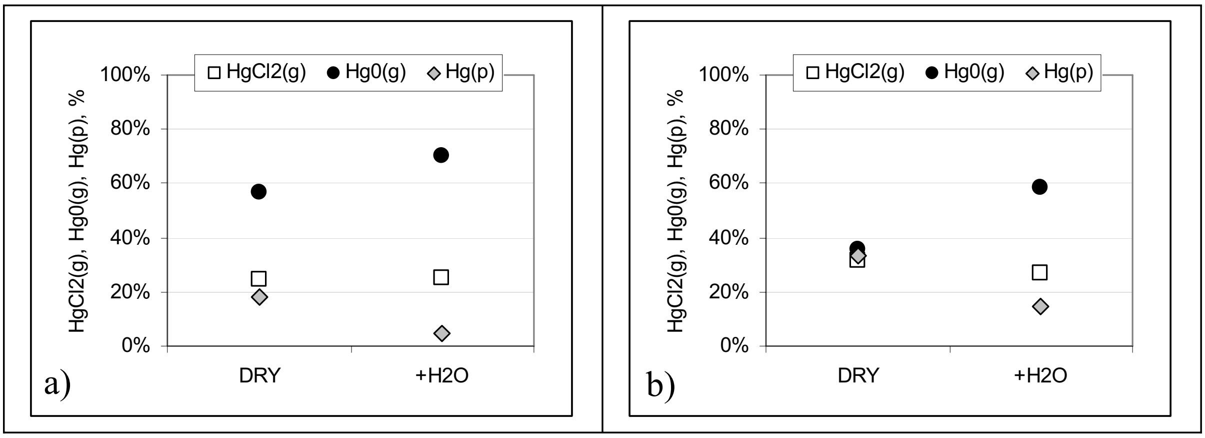


Fig. C-1/2: Influence of additional water on mercury speciation (staged) a) 1100°C b) 1300°C

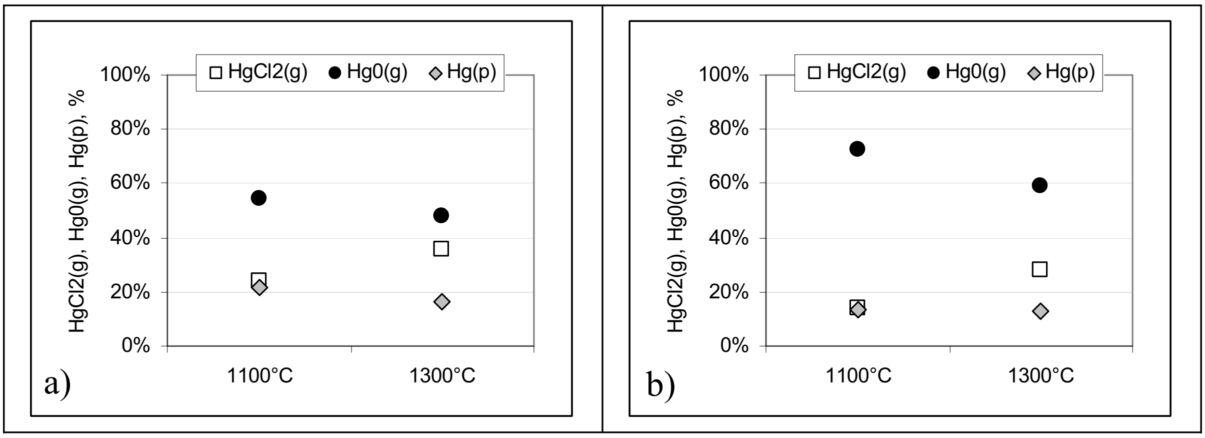


Fig. C-1/3: Influence of process temperature on mercury speciation (unstaged) a) dry b) + H_2O

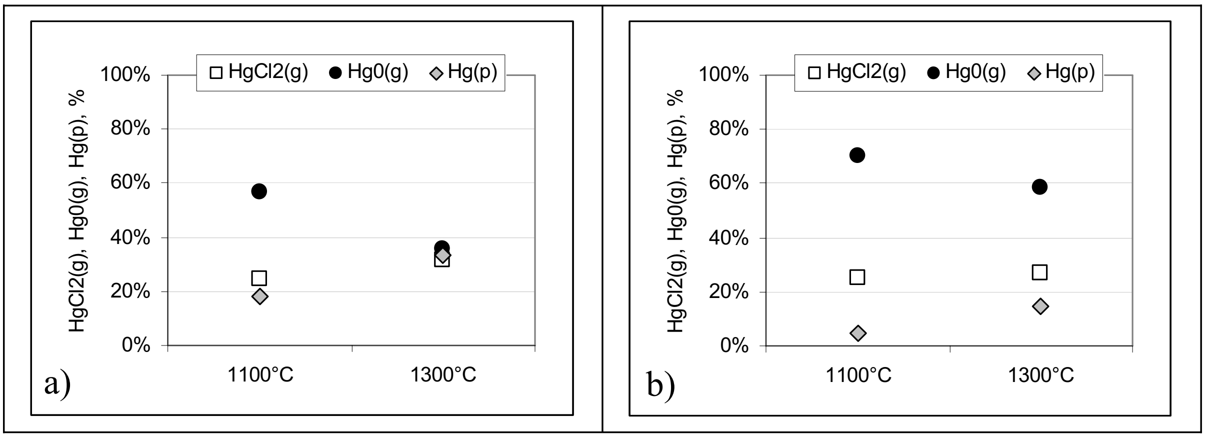


Fig. C-1/4: Influence of process temperature on mercury speciation (staged) a) dry b) + H_2O

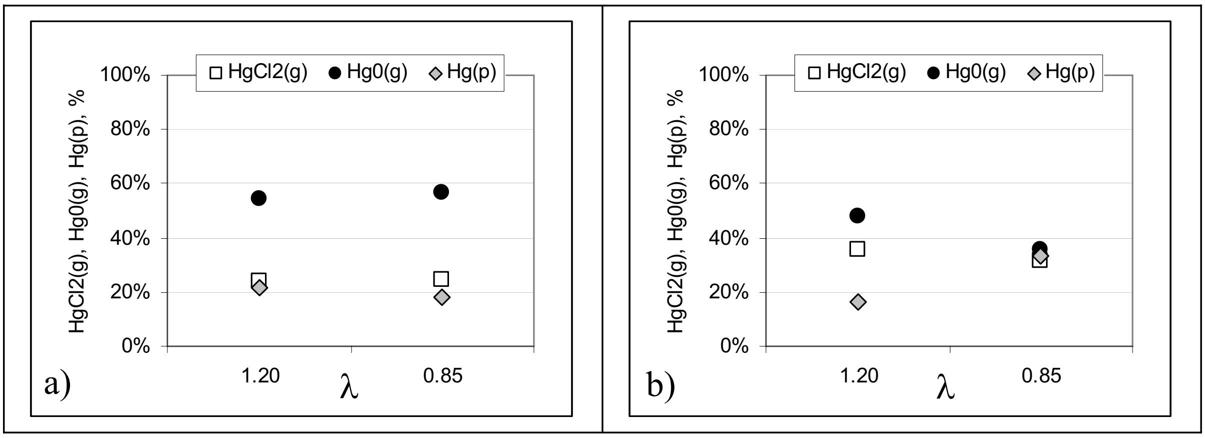


Fig. C-1/5: Influence of burner lambda on mercury speciation (dry) a) 1100°C b) 1300°C

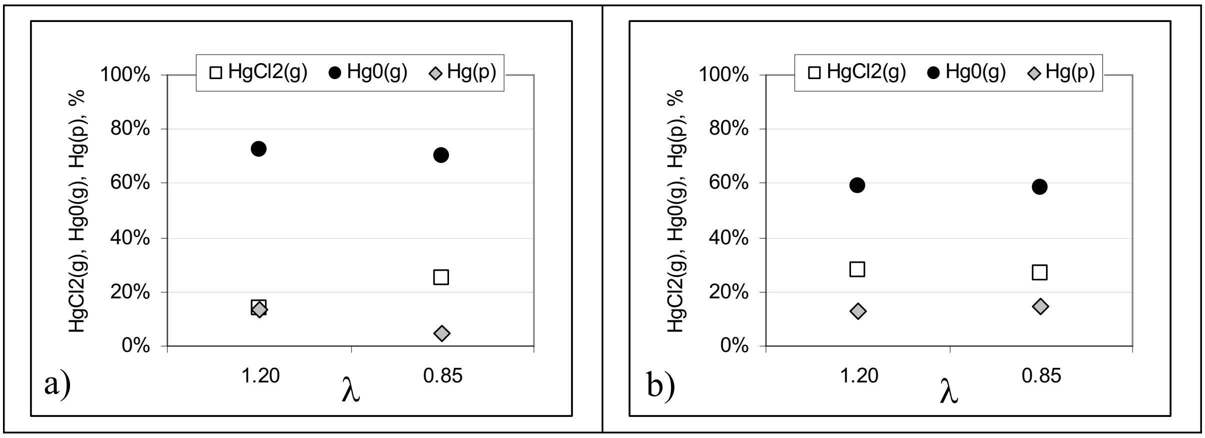


Fig. C-1/6: Influence of burner lambda on mercury speciation (+H_2O) a) 1100°C b) 1300°C

3.4 Influence of operational conditions on mercury speciation (bituminous B/1)

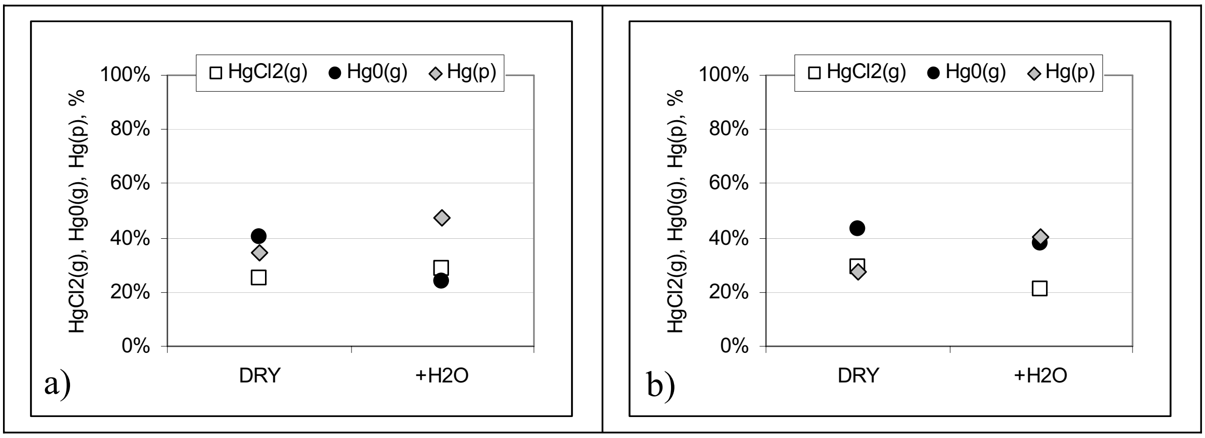


Fig. C-1/7: Influence of additional water on mercury speciation (unstaged) a) 1100°C b) 1300°C

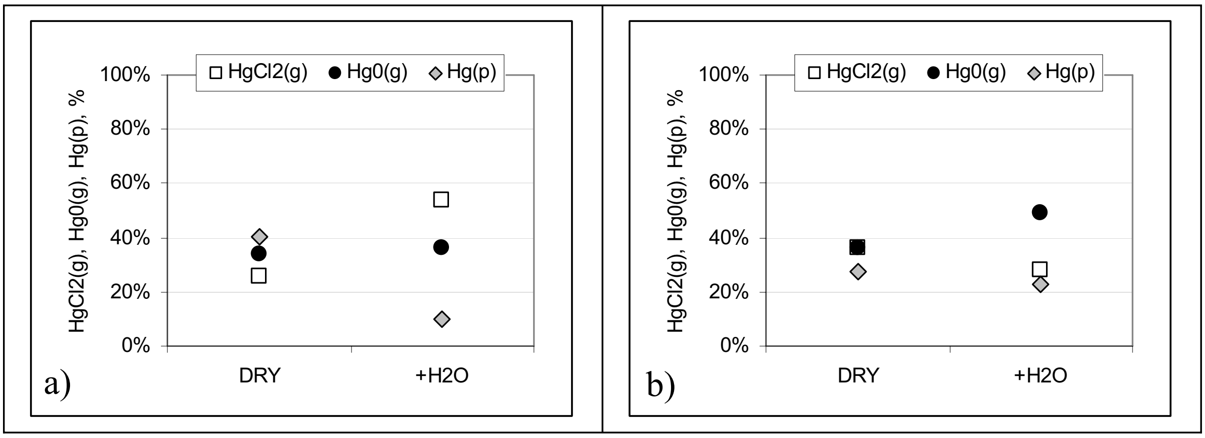


Fig. C-1/8: Influence of additional water on mercury speciation (staged) a) 1100°C b) 1300°C

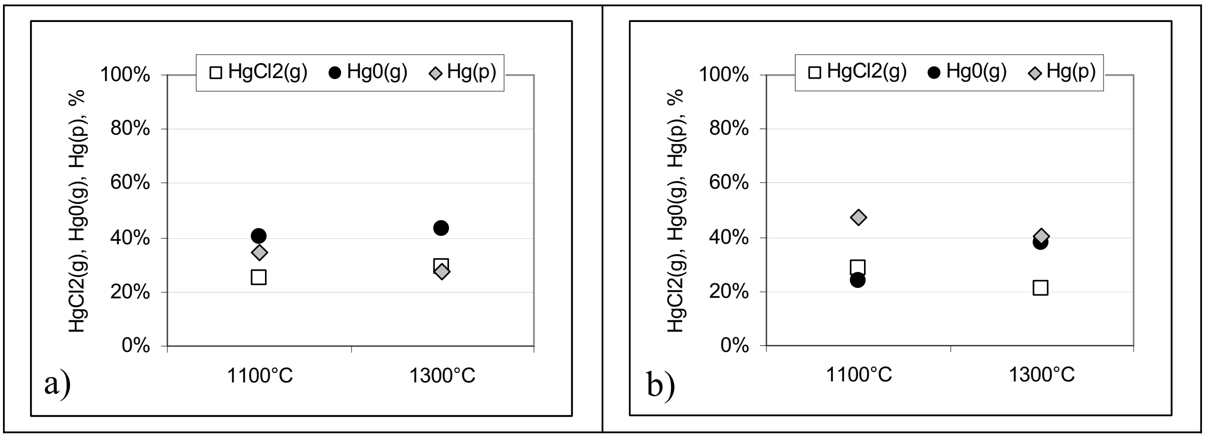


Fig. C-1/9: Influence of process temperature on mercury speciation (unstaged) a) dry b) + H_2O

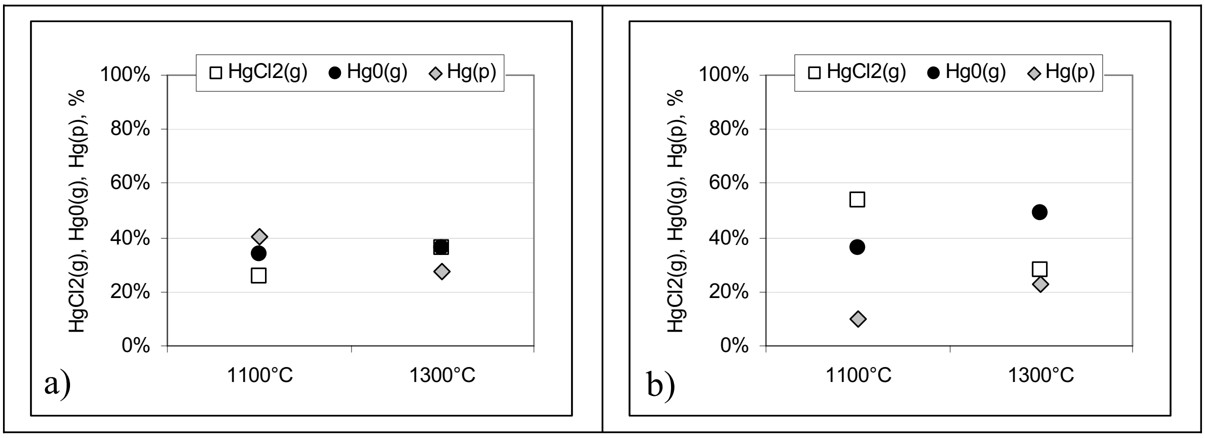


Fig. C-1/10: Influence of process temperature on mercury speciation (staged) a) dry b) + H_2O

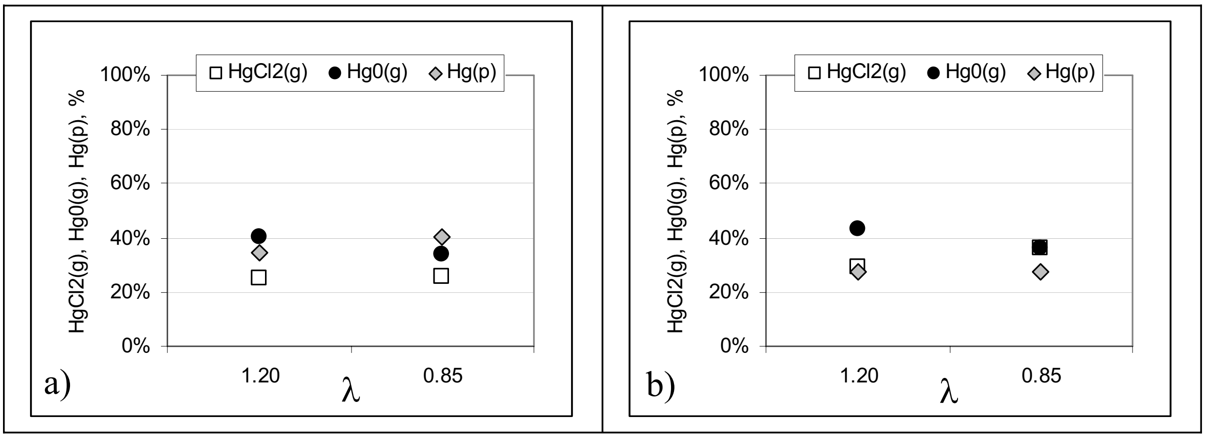


Fig. C-1/11: Influence of burner lambda on mercury speciation (dry) a) 1100°C b) 1300°C

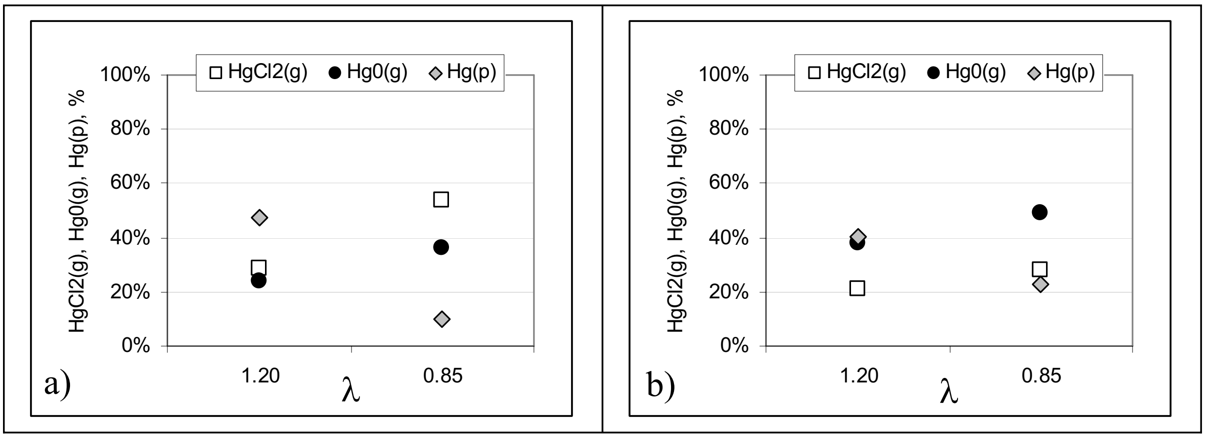


Fig. C-1/12: Influence of burner lambda on mercury speciation (+H_2O) a) 1100°C b) 1300°C

3.5 Influence of additional chlorine on mercury speciation (lignite L/1)

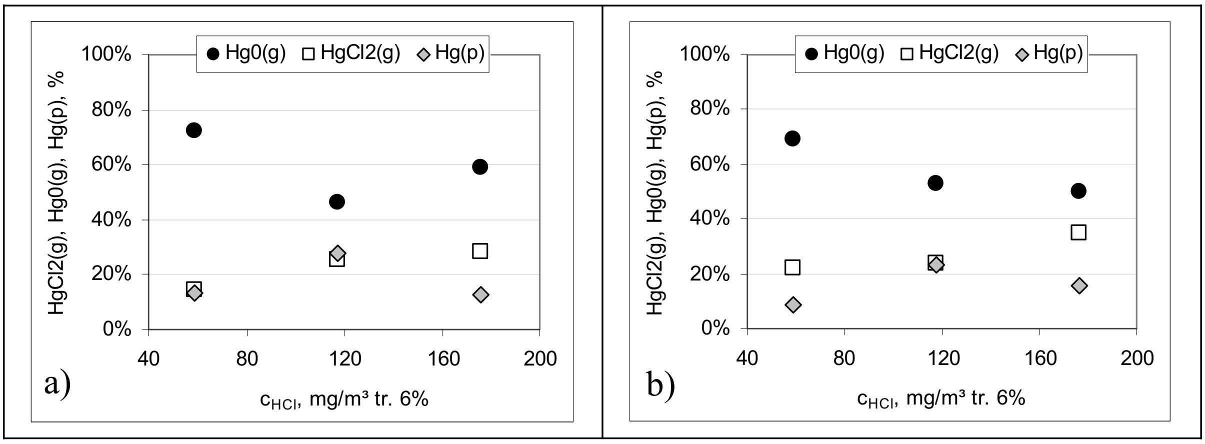


Fig. C-1/13: Influence of additional chlorine on mercury speciation a) V1 b) V2

3.6 Fate of additional mercury in the integrated process (lignite L/1)

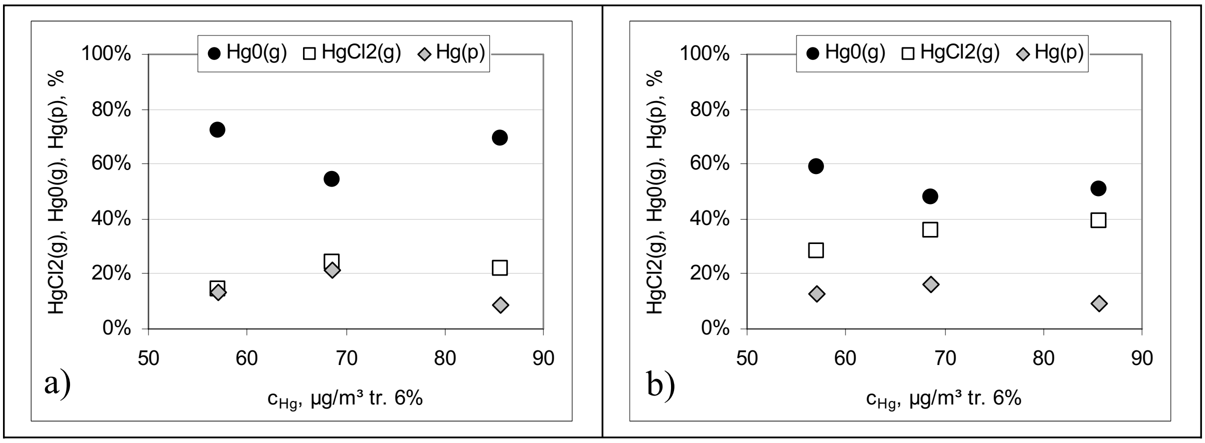


Fig. C-1/14: Lignite L/1, unstaged combustion a) 1100°C b) 1300°C

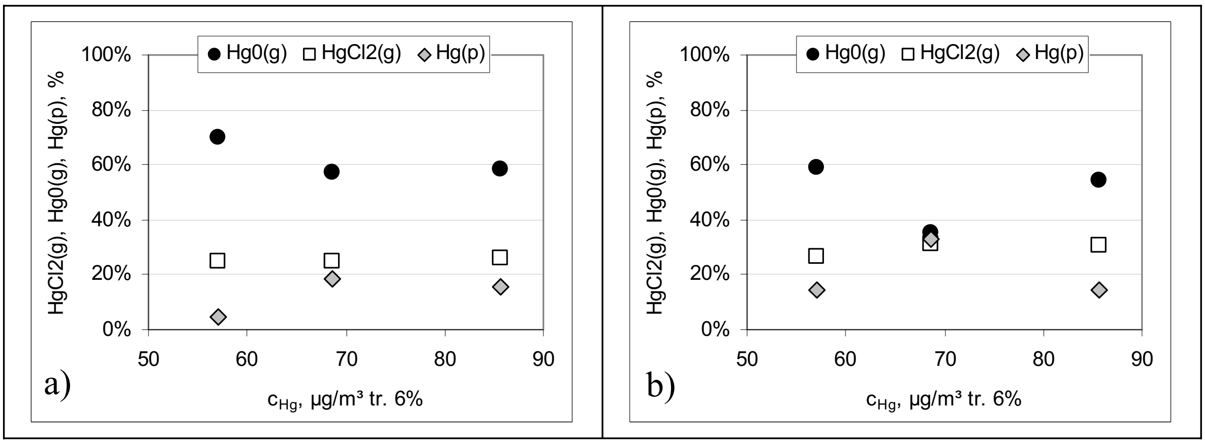


Fig. C-1/15: Lignite L/1, staged combustion a) 1100°C b) 1300°C

4. Results for particle-bound elements

4.1 Results of the trials with the Polish lignite L/1

Table C-1/21: Lignite L/1, 1100°C, λ = 1.20

V	Hg	Cd	Zn	Ni	As	Cu	Cr	Pb
-	mg/kg	Mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	Mg/kg	mg/kg
001	0.23	0.37	115	59	12	55	118	71
111	0.15	0.30	113	64	15	53	95	88
113	0.14	0.28	115	68	14	52	94	85

Table C-1/22: Lignite L/1, 1100°C, λ = 0.85

V	Hg	Cd	Zn	Ni	As	Cu	Cr	Pb
-	mg/kg	Mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg
003	0.18	0.81	161	62	25	58	135	108
131	0.05	0.42	138	64	24	50	101	106
133	0.26	0.39	124	66	23	51	94	▨

Table C-1/23: Lignite L/1, 1300°C, λ = 1.20

V	Hg	Cd	Zn	Ni	As	Cu	Cr	Pb
-	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg
311	0.10	0.49	84	79	15	59	109	93
312	0.10	0.46	82	60	13	61	105	87
314	0.10	0.42	80	61	12	75	110	79

Table C-1/24: Lignite L/1, 1300°C, λ = 0.85

V	Hg	Cd	Zn	Ni	As	Cu	Cr	Pb
-	mg/kg	Mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg
331	0.24	0.88	190	69	20	84	125	118
332	0.10	0.74	220	65	15	72	122	107
334	0.16	0.76	200	78	16	61	124	108

Table C-1/25: Lignite L/1, 1100°C, Hg = 0 µg/m³, HCl = 0, 400, 800 mg/m³

V	Hg	Cd	Zn	Ni	As	Cu	Cr	Pb
-	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg
111	0.15	0.30	113	64	15	53	95	88
221	0.26	0.42	110	79	13	59	139	98
231	0.10	0.29	76	68	11	73	126	69

Table C-1/26: Lignite L/1, 1100°C, Hg = 200 µg/m³, HCl = 0, 400, 800 mg/m³

V	Hg	Cd	Zn	Ni	As	Cu	Cr	Pb
-	mg/kg	Mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg
113	0.14	0.28	115	68	14	52	94	85
223	0.32	0.28	75	61	10	57	104	79
233	0.17	0.26	66	59	11	57	106	75

4.2 Results of the trials with the Colombian bituminous coal B/1

Table C-1/27: Bituminous coal B/1, 1100°C, λ = 1.20

V	Hg	Cd	Zn	Ni	As	Cu	Cr	Pb
-	mg/kg	mg/kg	mg/kg	mg/kg	Mg/kg	mg/kg	mg/kg	mg/kg
511	0.18	2.00	280	142	38	91	238	72
512	0.30	1.90	245	121	35	79	194	55
514	0.58	2.00	250	121	35	78	225	65

Table C-1/28: Bituminous coal B/1, 1100°C, λ = 0.85

V	Hg	Cd	Zn	Ni	As	Cu	Cr	Pb
-	Mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg
531	0.25	2.00	245	123	39	80	252	73
533	0.10	3.10	265	132	51	90	237	59
534	0.36	3.00	260	120	50	82	208	55

Table C-1/29: Bituminous coal B/1, 1300°C, λ = 1.20

V	Hg	Cd	Zn	Ni	As	Cu	Cr	Pb
-	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg
411	0.10	1.80	220	111	32	104	204	175
412	0.29	1.80	255	112	32	81	239	165
414	0.10	2.00	265	113	33	83	245	145

Table C-1/30: Bituminous coal B/1, 1300°C, λ = 0.85

V	Hg	Cd	Zn	Ni	As	Cu	Cr	Pb
-	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg
431	0.10	3.10	475	112	44	78	214	139
432	0.10	2.60	410	123	39	78	257	152
434	0.42	3.40	480	132	46	84	223	164

4.3 Influence of flue gas integration on trace elements in the fly ash

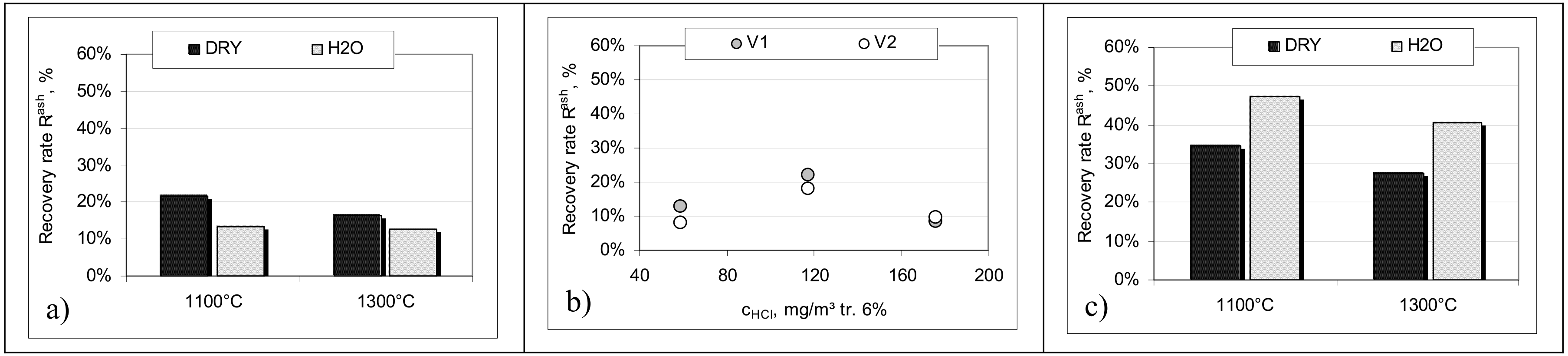


Fig. C-1/16: Mercury (Hg) a) Lignite L/1-H_2O b) Lignite L/1-HCl c) Bituminous coal B/1-H_2O

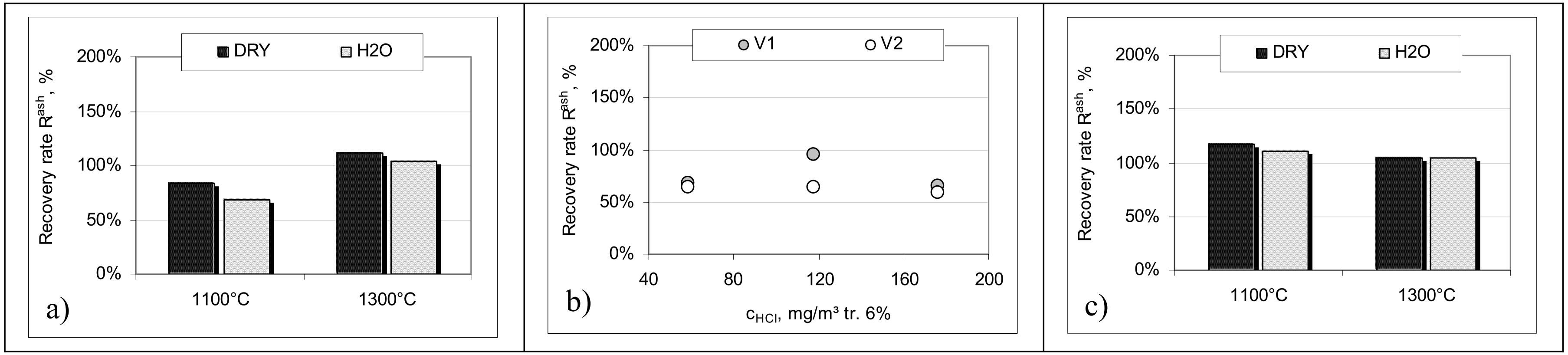


Fig. C-1/17: Cadmium (Cd) a) Lignite L/1-H_2O b) Lignite L/1-HCl c) Bituminous coal B/1-H_2O

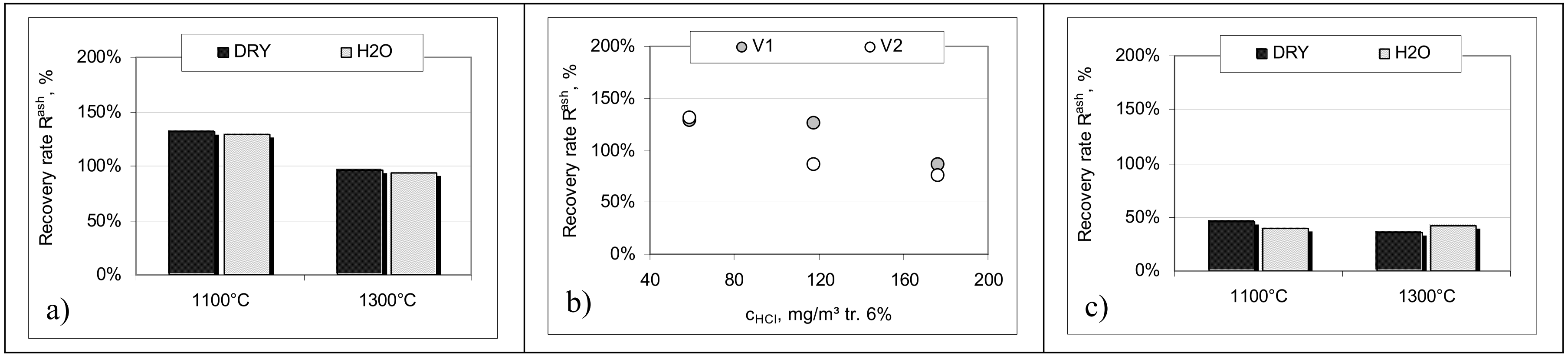


Fig. C-1/18: Zinc (Zn) a) Lignite L/1-H_2O b) Lignite L/1-HCl c) Bituminous coal B/1-H_2O

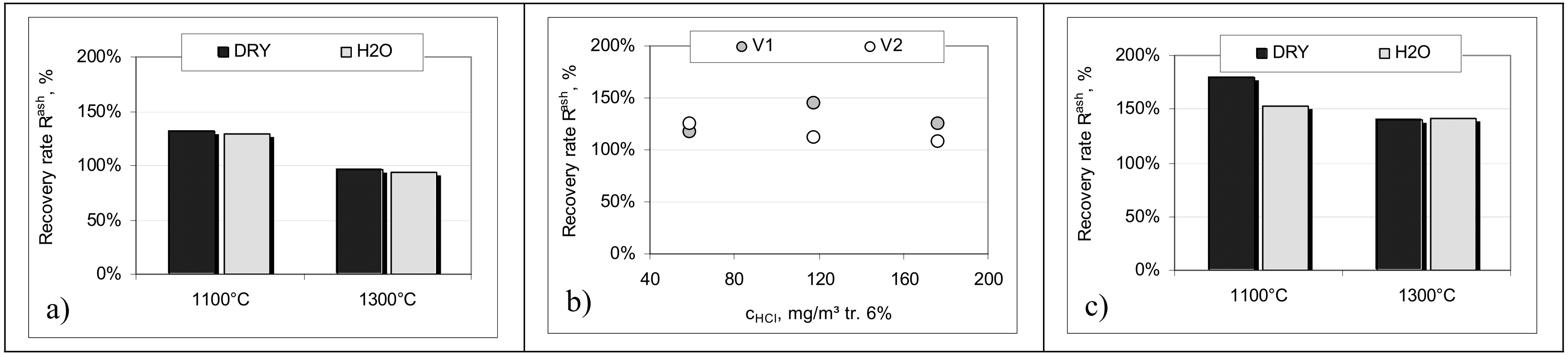


Fig. C-1/19: Nickel (Ni) a) Lignite L/1-H_2O b) Lignite L/1-HCl c) Bituminous coal B/1-H_2O

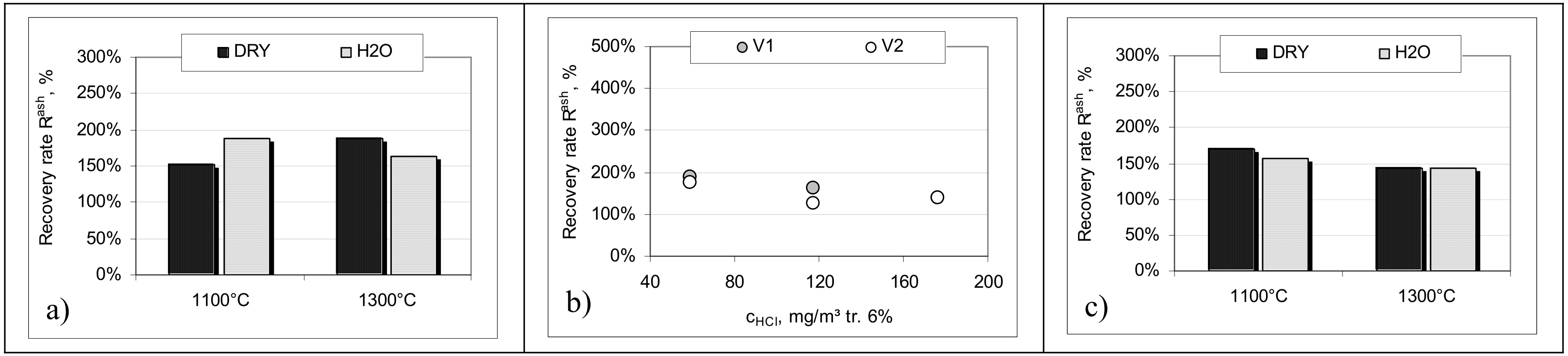


Fig. C-1/20: Arsenic (As) a) Lignite L/1-H_2O b) Lignite L/1-HCl c) Bituminous coal B/1-H_2O

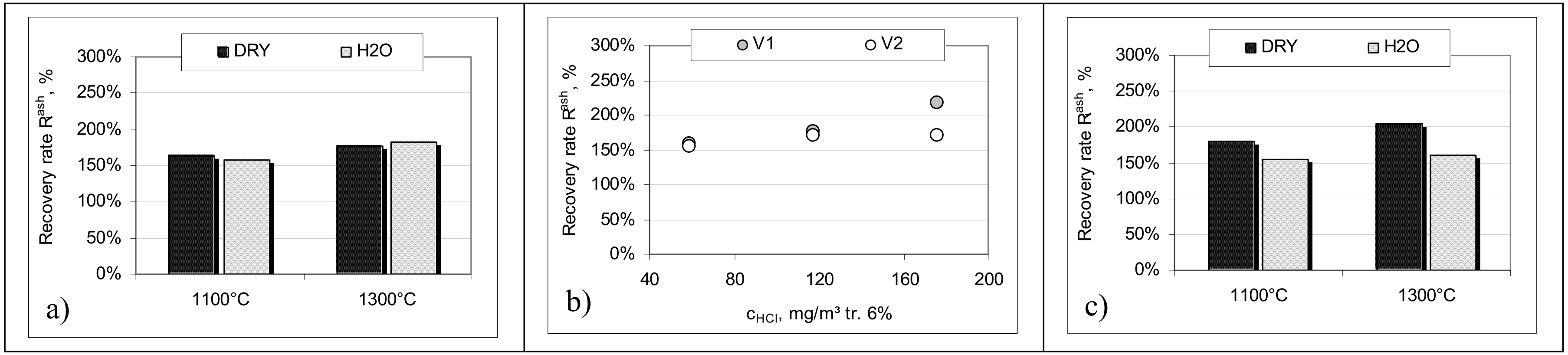


Fig. C-1/21: Copper (Cu) a) Lignite L/1-H_2O b) Lignite L/1-HCl c) Bituminous coal B/1-H_2O

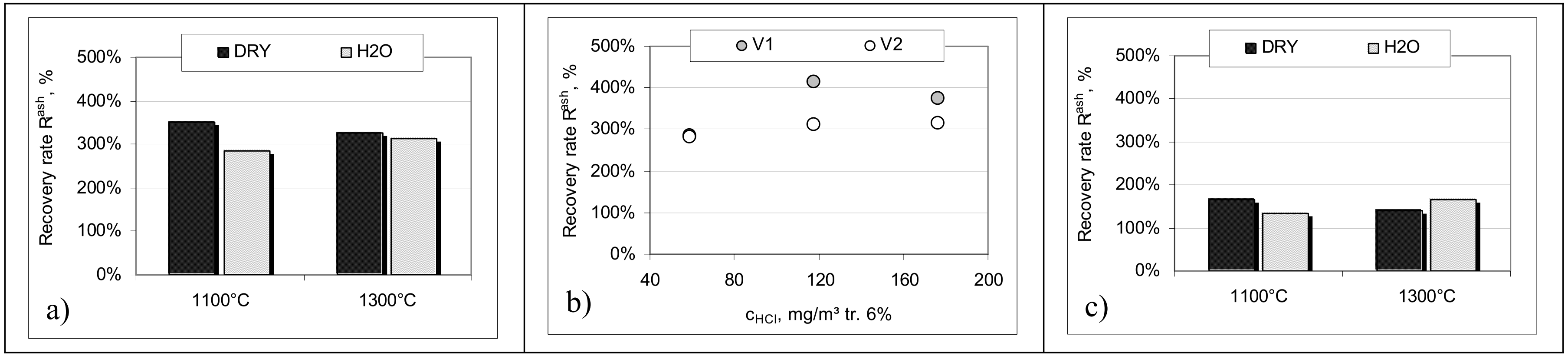


Fig. C-1/22: Chromium (Cr) a) Lignite L/1-H_2O b) Lignite L/1-HCl c) Bituminous coal B/1-H_2O

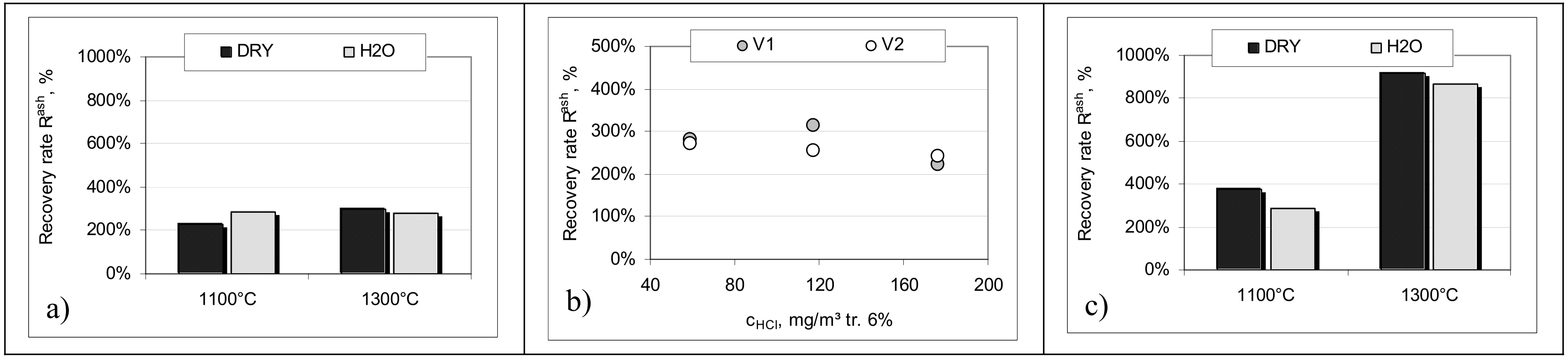


Fig. C-1/23: Lead (Pb) a) Lignite L/1-H_2O b) Lignite L/1-HCl c) Bituminous coal B/1-H_2O

Appendix C-2 : Trace elements SRF co-combustion

1. Combustion settings

1.1 Combustion settings KSVA

Table C-2/1: Combustion settings bituminous coal B/2 – SRF/PRR

Coal, kg/h	P, kW	SRF share	SRF, kW	SRF, g/h	Ash, kg/h	P, kW
35.00	299.79	0.0%	0.00	0	2.87	299.79
33.25	284.80	5.0%	14.99	2367	2.93	299.79
31.50	269.81	10.0%	29.98	4734	2.99	299.79

Table C-2/2: Combustion settings bituminous coal B/3 – SRF/PRR

Coal, kg/h	P, kW	SRF share	SRF, kW	SRF, g/h	Aash, kg/h	P, kW
33.50	301.85	0.0%	0.00	0	1.81	301.85
31.83	286.76	5.0%	15.09	2383	1.93	301.85
30.15	271.67	10.0%	30.19	4767	2.04	301.85

1.2 Combustion settings BTS

Table C-2/3: Combustion settings bituminous coal B/1 – SRF

Coal, kg/h	P, kW	SRF share	SRF, kW	SRF, g/h	Ash, kg/h	P, kW
0.800	5.92	0.0%	0.00	0	0.12	5.92
0.768	5.69	5.0%	0.24	42	0.12	5.92
0.736	5.45	10.0%	0.47	85	0.12	5.92

Table C-2/4: Combustion settings lignite L/3 – SRF

Coal, kg/h	P, kW	SRF share	SRF, kW	SRF, g/h	Ash, kg/h	P, kW
1.200	5.99	0.0%	0.00	0	0.09	5.99
1.152	5.75	5.0%	0.24	43	0.09	5.99
1.104	5.51	10.0%	0.48	86	0.10	5.99

2. Gaseous- and particle-bound mercury

2.1 Co-combustion trials BTS

Table C-2/5: SRF/04-B/1 (Colombian bituminous coal B/1)

V	c_{HgCl2}^{FG}	c_{Hg0}^{FG}	c_{Hg}^{FG}	c_{Hg}^{ash}	R_{Hg}^{tot}	$HgCl_2(g)$	$Hg^0(g)$	Hg(p)
-	µg/m³	µg/m³	µg/m³	µg/kg	%	%	%	%
0%	1.53	0.34	1.87	70	21.4%	47.6%	10.6%	41.9%
5%	▨	▨	▨	310	▨	▨	▨	▨
10%	2.17	0.71	2.88	80	28.9%	50.0%	16.4%	33.6%

Table C-2/6: SRF/04-L/3 (German lignite L/3)

V	c_{HgCl2}^{FG}	c_{Hg0}^{FG}	c_{Hg}^{FG}	c_{Hg}^{ash}	R_{Hg}^{tot}	$HgCl_2(g)$	$Hg^0(g)$	Hg(p)
-	µg/m³	µg/m³	µg/m³	µg/kg	%	%	%	%
0%	3.98	0.28	4.26	80	25.6%	72.3%	5.1%	22.7%
5%	3.44	0.38	3.82	140	28.5%	56.6%	6.3%	37.1%
10%	3.62	0.27	3.89	150	30.0%	57.2%	4.3%	38.5%

Table C-2/7: SRF/05-L/3 (German lignite L/3)

V	c_{HgCl2}^{FG}	c_{Hg0}^{FG}	c_{Hg}^{FG}	c_{Hg}^{ash}	R_{Hg}^{tot}	$HgCl_2(g)$	$Hg^0(g)$	Hg(p)
-	µg/m³	µg/m³	µg/m³	µg/kg	%	%	%	%
0%	3.98	0.28	4.26	80	25.6%	72.3%	5.1%	22.7%
5%	3.91	0.62	4.53	70	26.0%	69.1%	11.0%	19.9%
10%	▨	▨	▨	190	▨	▨	▨	▨

2.2 Influence of co-combustion on mercury speciation (Lignite L/3)

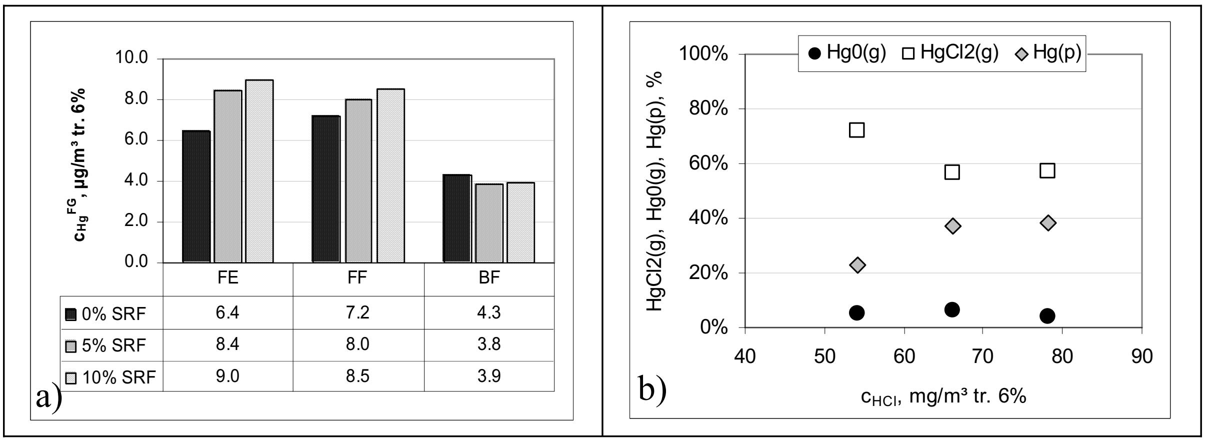


Fig. E-1/1: SRF/04-L/3 a) total mercury FE (furnace exit), FF (in front of filter), BF (behind filter) b) mercury speciation behind filter (BF)

2.3 Influence of chlorine concentration on (gaseous) mercury speciation (Lignite L/3)

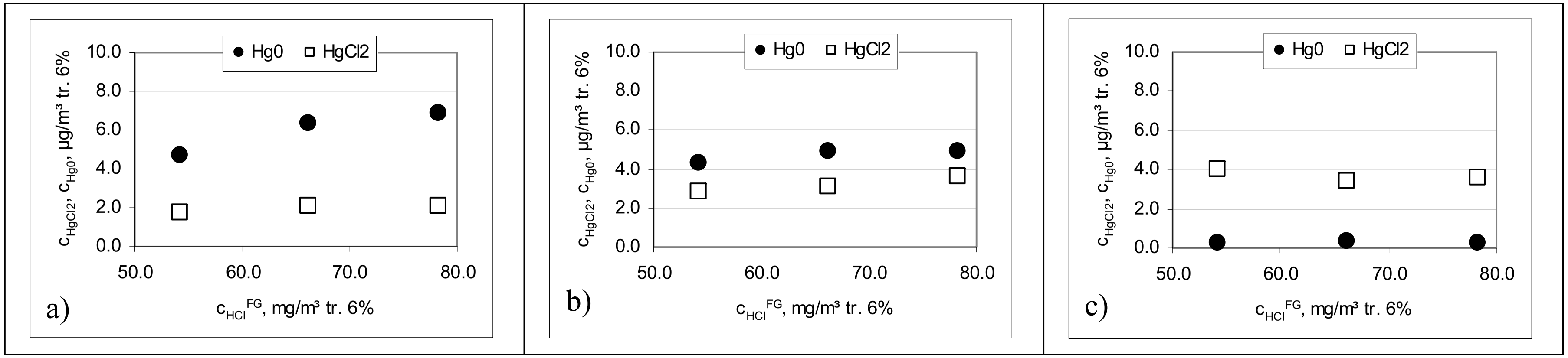


Fig. C-2/2: SRF/04-L/3 gaseous mercury, µg/m³ tr. 6% a) furnace exit (FE) b) in front of filter (FF) c) behind ash filter (BF)

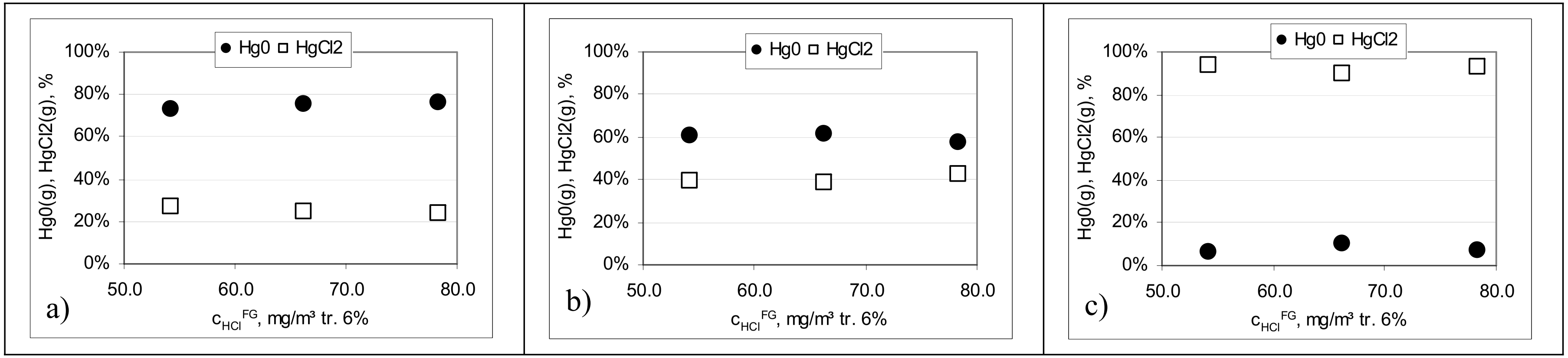


Fig. C-2/3: SRF/04-L/3 gaseous mercury, normalised a) furnace exit (FE) b) in front of filter (FF) c) behind ash filter (BF)

2.3 Influence of measurement location on (gaseous) mercury speciation (Lignite L/3)

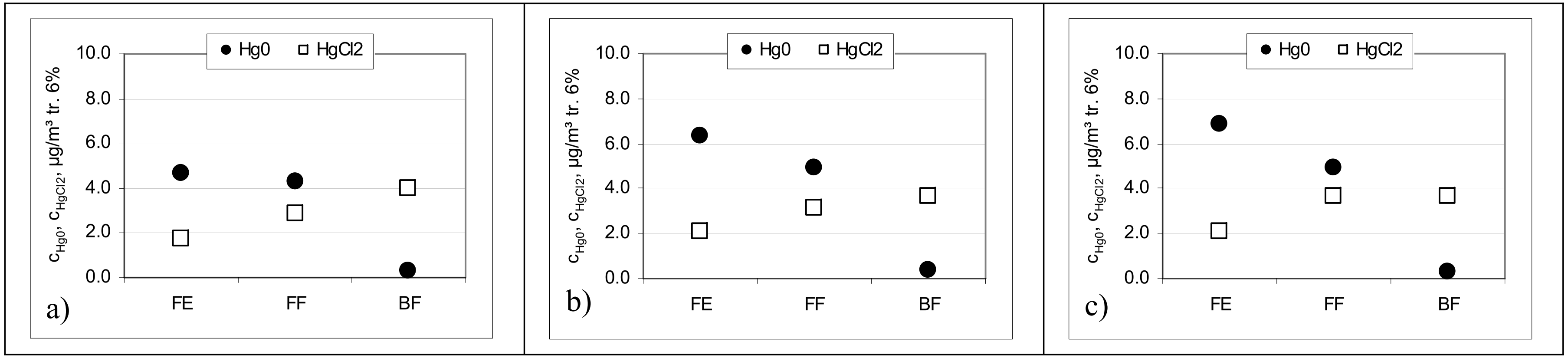


Fig. C-2/4: SRF/04-L/3 gaseous mercury, µg/m³ tr. 6% a) 0% thermal share b) 5% thermal share c) 10% thermal share

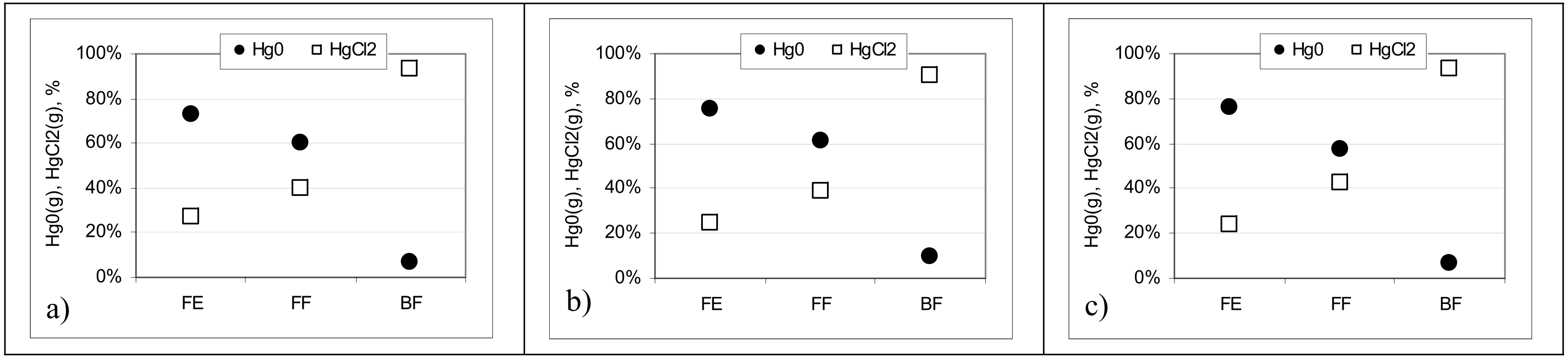


Fig. C-2/5: SRF/04-L/3 gaseous mercury, normalised a) 0% thermal share b) 5% thermal share c) 10% thermal share

Appendix D-1 : Legal limits

Table F-1/1: Comparison of emission limits (daily average for new plants)

		Directive 2001/80/EC			German 13. BImSchV			Directive 2000/76/EC	German 17.BImSchV
		50-100 MW	100-300 MW	>300 MW	50-100 MW	100-300 MW	>300 MW		
O_2	%	6			6			11 (6***)	11 (6***)
Dust	mg/m³	100 *		50 **	20	10		10	10
HCl	mg/m³	-			-			10	10
HF	mg/m³	-			-			1	1
SO_2	mg/m³	850	200		850	200		50	50
NO_x	mg/m³	400	200		400	200		200	200
CO	mg/m³	-			150	200		50	50
Hg	mg/m³	-			0.03			0.05	0.03
Cd+Tl	mg/m³	-			0.05			0.05	0.05
HM *	mg/m³	-			0.50			-	0.50
CP	mg/m³	-			0.05			-	0.05
TOC	mg/m³	-			-			10	10
PCDD/F	ng(TQ)/m³	-			0.10			0.10	0.10

* HM: Sum of (Sb+As+Pb+Cr+Co+Cu+Mn+Ni+V+Zn) CP (carcinogenic pollutants): As+Benzo(a)-pyren+Cd+Co+Cr * : <500 MW ** : >500 MW *** : co-combustion

Appendix D-1 : Legal limits

Table D-1/1: Requirements according to EN-450 for utilisation of fly ashes in concrete

Parameter	Legal limit	According to DIN
Loss on ignition	< 5% class A 2-7% class B 4-9% class C	EN 196-2 **
$SiO_2 + Al_2O_3 + Fe_2O_3$	> 70%	EN 196-2 **
Chloride (Cl^-)	< 0.1%	EN 196-2 **
Sulphuric anhydride (SO_3)	< 3%	EN 196-2 **
Free CaO	< 1% (< 2.5%) *	EN 197-1 ***
Reactive CaO	< 10%	EN 197-1 ***
Total alkaline content (as Na_2O)	< 5%	EN 196-2 **

* further investigations required if content of free CaO ranges between 1 and 2.5%

** DIN EN196-2: Chemical analysis of cement

*** DIN EN197-1: Composition, specifications and conformity criteria for common cements

Table D-1/2: Legal limits for eluates according to German *Versatzverordnung (VersatzV)*

Anorganic pollutants	In mg/l
Arsenic (As)	< 0.01
Lead (Pb)	< 0.025
Cadmium (Cd)	< 0.005
Chromium, total (Cr)	< 0.05
Chromate (Cr VI)	< 0.008
Copper (Cu)	< 0.05
Nickel (Ni)	< 0.05
Mercury (Hg)	< 0.001
Zinc (Zn)	< 0.5
Cyanide, total (CN^-)	< 0.05
Cyanide, volatile (CN^-)	< 0.01

Table D-1/3: Limits according German *Abfallablagerungsverordnung (AbfAblV)*

Parameter	Unit	Class 1	Class 2	Special wastes
Shear strength	kN/m²	> 25	> 25	> 25
Loss on ignition (LOI)	wt.-%	< 3	< 5	< 10
Extractable lipophile substances	wt.-%	< 0.4	< 0.8	< 4
Eluation criteria				
pH – value	-	5.5 - 13	5.5 - 13	4 - 13
Conductivity	µS/cm	< 10,000	< 50,000	< 100,000
TOC	mg/l	< 20	< 100	< 200
Phenols	mg/l	< 0.2	< 50	< 100
Arsenic	mg/l	< 0.2	< 0.5	< 1
Lead	mg/l	< 0.2	< 1	< 2
Cadmium	mg/l	< 0.05	< 0.1	< 0.5
Chromium	mg/l	< 0.05	< 0.1	< 0.5
Copper	mg/l	< 1	< 5	< 10
Nickel	mg/l	< 0.2	< 1	< 2
Mercury	mg/l	< 0.005	< 0.02	< 0.1
Zinc	mg/l	< 2	< 5	< 10
Fluoride	mg/l	< 5	< 25	< 50
Ammonia	mg/l	< 4	< 200	< 1,000
Chloride	mg/l	-	-	< 10,000
Cyanide	mg/l	< 0.1	< 0.5	< 1
Sulphate	mg/l	-	-	< 5.000
Nitrite	mg/l	-	-	< 30
AOX	mg/l	< 0.3	< 1.5	< 3
Water soluble residue	wt.-%	< 3	< 6	< 10

Appendix D-2: Analytical methods

a) Laboratory methods

Table F-2/1: Sample preparation and analytical methods

Analytical procedure	Method	Quantification limit
Sample preparation		
Preparation of lab ashes	DIN 51719	None
Preparation of eluates	DIN 38414 Part 4	None
Microwave digestion	DIN 22022 Part 1	None
Determination of physical properties		
Water content	DIN 51718	0.1 %
Ash content	DIN 51719	0.1 %
Volatile content	DIN 51720	0.1 %
Gross/net calorific value (GCV/NCV)	DIN 51900 Part 1+3	200 J/g
Sieve analysis	DIN 66165 Part 1+2	10 µg
Ash melting behaviour (following ASTM D 1857)	DIN ISO 51730	None
Determination of chemical properties		
Ultimate analysis (C, H, N, S)	following DIN 10649 following ASTM 3176	0.3 %
Chloride after bomb digestion	DIN 51727	0.03 %
Cl^-, F^-, SO_4^{2-} (Ion-chromatography)	DIN EN ISO 10304 Part 1+2	0.1 mg/l
Quantitative XRF analysis after $Li_2B_4O_7$ melting digestion (Si = 1.2 %, Al = 0.5 %, Fe = 0.4 %, Ca = 0.5 %, K = 0.3 %, Ti = 0.3 %, Na = 0.4 %, Mg = 0.2 %, P = 0.1 %, Mn = 0.01 %)	DIN 51729-10	0.01 - 1.2%
Heavy metals flame AAS	DIN 22022-3	0.1 mg/l
graphite AAS	DIN 22022-5	0.1 µg/l
Mercury Hybrid – AAS (FIMS)	DIN 22022-4	0.1 µg/l

b) In-situ gas measurement devices

Device	Company	Measured value	Range	Basic principle
Ultramat 21	Siemens	CO_2	0-100%	NDIR
Ultramat 23	Siemens	SO_2 CO O_2	0-10000 ppm 0-10000 ppm 0-20%	NDIR NDIR Fuel cell
CLD 822S	Eco Physics	NO NO_2	0-5000 ppm 0-5000 ppm	Chemilumineszenz (photoelectrical)
Binos 100	Rosemount	CO CO_2	0-1000 ppm 0-20%	NDIR NDIR
Binos 100	Rosemount	CO CO_2	0-10% 0-20%	NDIR NDIR
Oxynos 100	Rosemount	O_2	0-100%	Paramagnetic
S710	Sick Maihak	O_2	0-50%	Paramagnetic

c) Conversion of measured concentrations

$$CO\left[mg/m^3\right] = 1.250 \cdot CO\left[ppm\right] \tag{D-2.1}$$

$$NO_x\left[mg/m^3\right] = 2.054 \cdot NO_x\left[ppm\right] \tag{D-2.2}$$

$$SO_2\left[mg/m^3\right] = 2.930 \cdot SO_2\left[ppm\right] \tag{D-2.3}$$

d) Normalisation of gas concentrations to reference oxygen content

$$c_i(6\%) = c_i \cdot (21\% - 6\%)/(21\% - c_{O_2,meas}) \tag{D-2.4}$$

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