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From waste to raw material—the route from biomass to wood ash for cadmium and other heavy metals

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Abstract

Energetic utilization of biomass is considered an environmentally safe way of providing energy, especially for process heat and district- heating purposes. The main advantage of energy from biomass is the CO_2-neutrality of this energy-production process. However, this process produces a solid by-products, namely ash, that has to be considered.

This ash contains nutrients like calcium, potassium and phosphorus that should be recycled to forest or agricultural soils, thus closing not only the carbon cycle but also the fluxes of mineral materials caused by these technologies. The problem is, however, that besides nutrients, the ash also contains heavy metals. Cadmium poses a special risk to the use of wood ash in agriculture. It pollutes a large fraction of the ash generated in a biomass combustion plant, namely the cyclone fly-ash and, to an even higher degree, the filter fly-ash or (where flue gas condensation is installed) the condensation sludge.

A medium-term solution to the recycling of solid residues from biomass combustion is blending cyclone fly-ash and bottom ash and using the mixture in agriculture. Although a large part of nutrients might be recycled in this manner, care has to be taken of the relatively high amount of cadmium in this material.

A new technology currently under development takes advantage of the different temperatures in a biomass combustion plant. This technology enables concentration of cadmium (and other volatile heavy metals) in a very small portion of the whole ash flux from a plant and the concentrations of environmentally relevant substances in the remainder of the ash is kept low. In this manner, wood ash from the process industry or district heating systems might be transformed from waste to raw material for agricultural use.

Keywords: Biomass; Combustion; Wood ash; Cadmium; Heavy metal; Fertilizer

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1. Introduction

Biomass utilization for energy production is widely seen as part of the solution of pressing environmental problems, especially the accumulation of greenhouse gases in the atmosphere. In this respect, energy technologies can indeed contribute effectively since they close the carbon cycle, provided the generation of biomass by forestry is done in a sustainable way. However, although carbon and hydrogen form the bulk of the material flux going into a biomass combustion, other inorganic elements like calcium, potassium, magnesium, phosphorous and sodium, but also heavy metals like lead, zinc, cadmium, copper and nickel, participate in the material fluxes that are converted in thermal biomass utilization. They usually accumulate in the solid by-products, the ash. It is therefore necessary to look at the whole pattern of mass fluxes generated by thermal biomass utilization in order to evaluate its overall environmental impact. Only if the solid by-products can also be integrated into the biosphere, will energy from biomass really be a sustainable and clean technology that will contribute in the long run to the solution of our present ecological problems.

Usually, the bulk of the materials accumulated in ash from biomass combustion plants, namely the nutrients calcium, potassium, magnesium and phosphorus, should be recycled at the highest possible rate. They are elements needed for the growth of the plants that form the feedstock of biomass utilization. If it is possible to close these material cycles in addition to the carbon cycle, a truly sustainable technology for energy provision is possible. However, their recycling by using ash as fertilizers is impeded by accompanying heavy metals, most prominently cadmium. It must be the aim of any technology addressing the reintegration of solid by-products from thermal biomass utilization to concentrate the detrimental heavy metals in a tiny fraction of the ash in order to dispose of them and to recycle the rest of the ash safely to the biosphere in the form of secondary raw materials with fertilizing and liming effects. This necessitates knowledge both of the mass flows of these heavy metals within the whole system of combustion, heat utilization and ash precipitation and of the technological means available for improving the quality of the ash.

2. Where does cadmium in biomass come from?

It is important to state that the cadmium problem in biomass utilization is caused by the technology itself. It is imported via deposition onto the forest. The source of this deposition is the dissipation of cadmium by anthropogenic processes, most notably fossil energy use. In this context, clever biomass utilization can be seen as a way to reclaim, concentrate and finally dispose of cadmium that is dispersed by other processes. Fig. 1 gives a cadmium balance for Austrian conditions, including thermal biomass utilization by state of the art technologies ([1]). What can be seen from this figure is that the cadmium flow caused by biomass harvesting and biomass utilization is relatively small compared with the fluxes that are dissipated to and washed out from the forest ecosystem (the remainder of cadmium in this balance is accumulated in the top soil layers). The second lesson that can be drawn from this figure is that the dissipation of

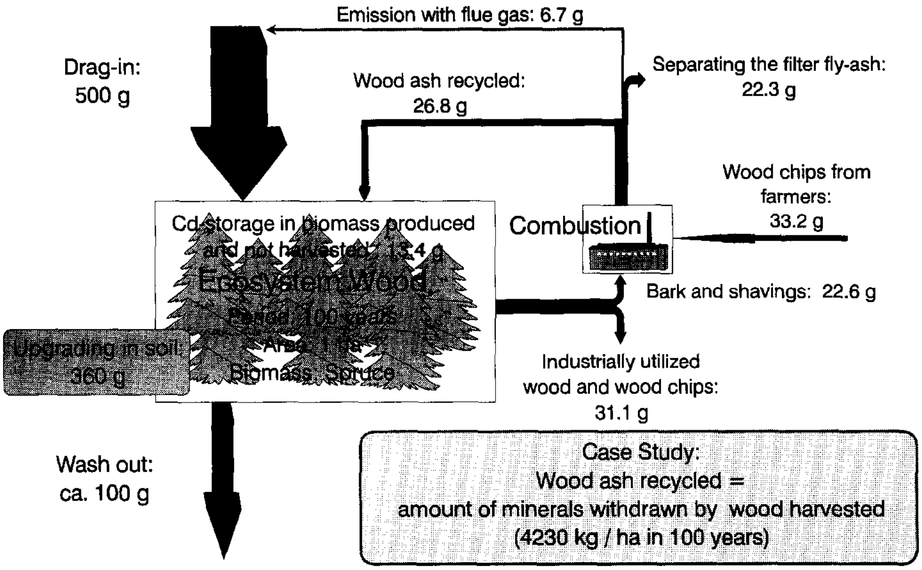

Fig. 1. Cadmium balance for the ecosystem wood under consideration of sustainable recycling of wood ash.

cadmium via dust emissions from the combustion plant into the air (and back to the forest ecosystem) is low. It is also clear from this figure that biomass can be seen as a concentration step for cadmium that can be used to purge a considerable amount of this heavy metal (4 to 5% of the total cadmium input in the forest ecosystem) in a controlled way. With careful handling of ash and with some technological improvements, thermal biomass utilization may emerge as a possible way to reduce the overall cadmium burden of the environment once the dissipation of the metal has been curbed.

The actual load of solid by-products generated by a biomass combustion plant varies significantly, depending on the raw materials used. The most important fuels used in combustion units based on woody biomass are bark, wood chips and sawdust. All these materials have a comparable net calorific value (between 2.3 $kWh\,kg^{-1}$ fresh substance for bark and 2.8 for wood chips). Their respective ash content, however, varies widely, from 0.5 wt% (dry basis) for sawdust to 1.6 wt% for wood chips and 6.0 wt% (or more) for bark ([2,3]). Although a certain dilution effect may be seen in the case of bark, which usually contains a considerable amount of inorganic impurities like sand or soil, for most metals the load per kWh primary energy input goes with the ash content of the fuel.

3. The fate of cadmium in the combustion plant

In a biomass combustion plant ash will occur at different steps of the process (see Fig. 2 and Table 1). The relative masses of these flows differ with the kind of fuel used and the technology applied. For bark, bottom ash will typically constitute 75–85 wt% of the total ash, cyclone fly-ash will contribute 15–25% and a relatively small amount

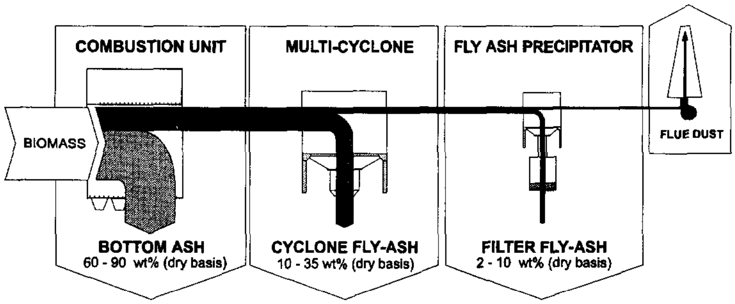

Fig. 2. Ash fractions produced in biomass combustion plants.

(1–4%) will be filter fly-ash. For wood chips as fuel, the distribution between the different types of ash will be comparable, however with wider margins. Sawdust combustion will, however, show a different picture, since these plants usually use another technology, namely underfeed stokers or direct-fired plants. In this case, bottom ash will be reduced (to 20–30%) with the main ash flow going to cyclone fly-ash (55–65%) and a considerable amount of filter fly-ash (10–15%) ([4,2]).

The important point in this respect is that the quality of the ash varies considerably in these different flows. Combustion acts like a thermodynamic separation process for the different inorganic materials contained in the fuel. Metals with low volatility, like nickel, chromium and vanadium will concentrate in the bottom ash and/or the cyclone fly-ash (see Table 2). The same is true for the main nutrients calcium, magnesium and phosphorus (see Table 3). Potassium is more volatile than the other nutrients but the major amount (about 85%) of this element is also found in the bottom ash and the cyclone fly-ash.

Most environmentally relevant metals like cadmium, zinc, lead and mercury will concentrate in the cyclone fly-ash and especially in the filter fly-ash. The key to this phenomenon is obviously the temperature at which these ash flows are precipitated. The bottom ash leaves the combustion chamber at temperatures between 600 and 1000°C, which keeps it virtually free of volatile heavy metals that are, coincidentally, also the environmentally most important ones.

In most biomass combustion plants the cyclones are installed after the heat exchanger units. Hence, they work at low temperatures, usually between 140 and 200°C. Temperatures at which filter fly-ash (or, in plants equipped with flue gas condensation, the condensation sludge) is precipitated are again lower than those in cyclones (40°C in flue

Table 1
Amount of the different ash fractions as a percentage (wt%) of the total amount of ash

Biomass fuel/ash fraction	Bark	Wood chips	Sawdust
Bottom ash	75–85	70–90	20–30
Cyclone fly-ash	15–25	10–30	55–65
Filter fly-ash	2–4	3–6	10–15

Table 2
Average distribution of highly volatile and least volatile heavy metals among the different ash fractions

Ash fraction/element	Bottom ash	Cyclone fly-ash (wt%)	Filter fly-ash
Highly volatile heavy metals			
Zn	11.1	43.8	45.1
Pb	9.8	35.4	54.8
Cd	3.4	54.0	42.7
Hg	2.6	12.9	84.5
Least volatile heavy metals			
Co	51.9	40.3	7.8
Ni	52.2	41.1	6.8
Cr	53.8	34.8	11.4
V	53.1	41.5	5.4

gas condensation units and between 100 and 140°C in electrostatic precipitators or fibrous filters).

At these temperatures desublimation and condensation of volatile metals and their compounds takes place or is already completed ([5–9]). Thus, the concentrations of these substances in these fractions tend to increase. In the case of cadmium, concentration in the cyclone fly-ash may be higher by a factor 10 to 20 and in the filter fly-ash by a factor of up to 200 compared with the bottom ash.

Besides the temperature, the gaseous atmosphere around the ash particles (oxidizing or reducing) and the surface of the fly-ash (the particle size) also seem to influence the precipitation of heavy metals. But, according to the current state of research, these influences only become important if the flue gas temperature remains under 700°C. Nevertheless, the gaseous atmosphere seems to play an important role for the sublimation or volatilization of heavy metals by chemical reactions in the bed of embers. A reducing atmosphere around the burning fuel particles seems to enforce and accelerate heavy metal volatilization because the formation of metal oxides, which are less volatile, is constrained due to lack of oxygen. Consequently, an understoichiometric atmosphere in the primary combustion zone will decrease the amount of heavy metals in the bottom ash.

Table 4 shows the amounts of environmentally relevant metals that leave a combustion plant with the different ash fractions. These data represent the results of a number of test runs and measurements that have been carried out in a typical Austrian biomass

Table 3
Average distribution of plant nutrients among the different ash fractions

Ash fraction/element	Bottom ash	Cyclone fly-ash (wt%)	Filter fly- ash
Ca	51.5	40.5	8.0
Mg	57.8	35.3	6.9
K	41.9	41.7	16.3
P	48.9	41.1	10.0
Na	49.5	41.0	9.5

Table 4
Measured distribution of the inorganic elements among the different ash fractions in a biomass combustion plant. *Explanations*: fuel: mixture of bark and wood chips; combustion technology: moving grate; specific plant capacity: 4 MW_{th}; cyclone fly-ash precipitation in a multi-cyclone working at about 180°C; filter fly-ash precipitation in a flue gas condensation unit working at about 40°C; ash analyses: pressurized acid digestion employing a special method for ashes that guarantees a complete and correct element detection ([10]) followed by ICP/OES resp. AAS

Element	Total amount (sum of all ash fractions)	Distribution amoung the different ash fractions			
	$mg\,kg^{-1}$ (dry basis)	Bottom ash (%)	Cyclone fly-ash (%)	Filter flyash (%)	Flue dust (%)
Si	78000	73.4	21.7	3.8	1.1
Ca	339000	66.1	29.0	3.8	1.1
Mg	28300	69.5	26.2	3.3	1.0
K	42900	55.6	35.2	7.1	2.1
Na	3300	60.7	22.4	13.0	3.9
P	10100	66.3	26.0	5.9	1.7
Al	24100	75.8	20.5	2.9	0.9
S	11300	25.6	51.8	17.5	5.2
Cl	3800	7.2	63.5	22.6	6.7
Fe	16700	70.4	26.6	2.3	0.7
Mn	19000	64.7	30.0	4.1	1.2
Cu	237.6	61.9	21.7	12.6	3.7
Zn	2094.5	23.2	45.2	24.4	7.2
Co	14.2	68.2	26.1	4.4	1.3
Mo	3.2	59.9	28.2	9.2	2.7
As	5.8	51.6	32.2	12.5	3.7
Ni	46.2	58.7	31.5	7.6	2.3
Cr	257.6	72.6	19.7	5.9	1.8
Pb	65.9	11.7	34.2	41.7	12.4
Cd	18.2	6.4	47.0	36.0	10.7
V	45.1	71.5	22.8	4.4	1.3
Hg	0.4	0.8	7.6	70.7	20.9
Percentage of the different ash fractions of the total amount of ash (wt%)		64.5	28.5	5.4	1.6

combustion plant that supplies a whole village with district heat and warm water (over 220 such plants are working in Austria at the moment). It can be seen that the relatively small fraction of the filter fly-ash (or condensate sludge) acts as a sink for many of the heavy metals, especially cadmium. Up to 36% of the cadmium entering the plant via the biomass fuel ends up in just 5.4% of the whole ash flux.

The consequence of this separation effect within biomass combustion plants is that the different ash fractions are differently suited for utilization as a secondary raw material with fertilizing and liming effects in forestry and agriculture. There is no doubt that bottom ash may be recycled. Cyclone fly-ash as such is already polluted to a degree that does not allow its use as a soil-improving agent. It may only be used in a mixture with the bottom ash according to the plant specific ash production ratio. Filter fly-ash (or condensation sludge) is in any case a highly polluted hazardous waste that has to be disposed of in a safe way or that could even be of interest for industrial heavy metal recovery.

4. Strategies for minimization of hazardous waste from biomass combustion plants

The main problem from the point of view of hazardous waste management of solid by-products from biomass combustion plants is the cyclone fly-ash. On the one hand, it contains a large amount of nutrients—40 wt% of calcium, 41% of phosphorus and sodium, 42% of potassium and 35% of manganese contained in the fuel (based on a state-of-the-art biomass combustion plant) will end up in the cyclone fly-ash. From the point of view of sustainability a loss of these nutrients is a serious disadvantage. Disposal of the cyclone fly-ash as hazardous waste opens a material cycle that might be closed and necessitates the use of mineral fertilizers to cover the loss.

On the other hand, disposal of cyclone fly-ash is a considerable economic burden to biomass utilization. A rough estimate on the basis of Austrian experience shows that the annual production of cyclone fly-ash is about 5 t per MW power installed. Given the costs for disposal of hazardous waste, this amounts to a considerable part of the operating costs of biomass district heating plants. This economic problem is even more serious as biomass utilization is in a fierce competition with other forms of energy provision on a fossil fuel base.

A quick and obvious solution to this dilemma is to mix bottom ash and cyclone fly-ash in the same ratio as they are produced in the plant. This, however, necessitates additional equipment in the plant to blend the two fractions and to condition the mixture for use by farming equipment, mainly by sieving of large particles that may impede distribution on fields or in forests with conventional manure spreaders.

Table 5 shows the average concentrations of environmentally relevant metals in a mixture of bottom and cyclone fly-ash, based on thorough studies of 32 Austrian biomass district heating plants using different biomass fuels and equipped with different

Table 5
Average heavy metal concentrations (mg kg^{-1}; dry basis) in mixtures of bottom ash and cyclone fly-ash in comparison with limiting values for sewage sludge application in Austria

Parameter/ element	Bark ash	Wood chip ash	Sawdust ash	Limiting value
Cu	93.4	133.1	202.0	500
Zn	738.4	347.6	1434.2	2000
Co	21.4	14.1	15.0	100
Mo	3.0	1.8	4.0	20
As	7.4	7.8	5.3	[20]
Ni	67.2	52.3	52.3	100
Cr	124.6	67.9	228.3	500
Pb	24.9	23.3	38.9	500
Cd	5.4	4.1	18.1	10

combustion technologies. The concentrations are contrasted with Austrian limiting values for the use of sewage sludge on agricultural fields. Although the standards used here might differ from country to country, the problem of this strategy is clearly visible. For most heavy metals there is a considerable margin of security between the metal concentrations in the blended ash and the environmental standards for distribution on fields. The sole exception is the concentration of cadmium; this may exceed the limiting value (especially in ashes from sawdust-fired plants that produce high amounts of cyclone fly-ash). A possible problem might also arise with nickel and zinc, for which the concentrations in the ash come closer to the environmental standards (at least in Austria).

A more objective picture gives the comparison between the concentrations of environmentally relevant metals in the blended ash and their respective concentrations in the top soil layers. In this case (and for Austrian average values) the concentration of zinc in the ash exceeds the recommended value for soils by roughly a factor of 2.5, nickel and chromium will slightly exceed them and the cadmium concentration in the ash is almost three times higher than in top soil layers. All the other metal concentrations in the blended ash are below or even far below the concentrations in soils.

It is clear from these results that blending of cyclone fly-ash and bottom ash leads to a product that may be recycled to forests or agricultural fields with some care. A thorough study on its usefulness as a secondary raw material with fertilizing and liming effects in agriculture and forestry is currently under way in Austria. Preliminary results show that this strategy is feasible as long as the annual ash application per hectare is limited (up to $2000\,kg\,ha^{-1}$ on agricultural fields, up to $1000\,kg\,ha^{-1}$ on grassland), thus guaranteeing negligible accumulation of environmentally detrimental heavy metals especially cadmium. However, this strategy may only be seen as a temporary solution to the problem of reintegrating solid by-products of biomass combustion in natural cycles. The results make it clear, however, that new technological solutions have to be developed in order to make the thermal utilization of biomass a sustainable long-term solution to environmental problems.

5. Technological approaches to minimize cadmium load in wood ash

Any technological approach towards improving the quality of residues from biomass combustion plants must concentrate on the following features:

- Increasing the part of ash that can be used in agriculture in a sustainable way by reducing the concentration of cadmium (and other environmentally relevant volatile heavy metals) in the respective ash fractions (especially in the cyclone fly-ash)
- Concentration of as much cadmium as possible in an as small an ash fraction as possible in order to reclaim it from disposed immersions and hence reduce the load of cadmium in the soil.

Besides the obvious advantage of making the process of thermal biomass utilization sustainable, an effective technology along these lines could also be used to clean contaminated soils from heavy metals. This could be done by planting special clones of Salix on these areas and subsequently using the biomass produced as fuel. Comprehen-

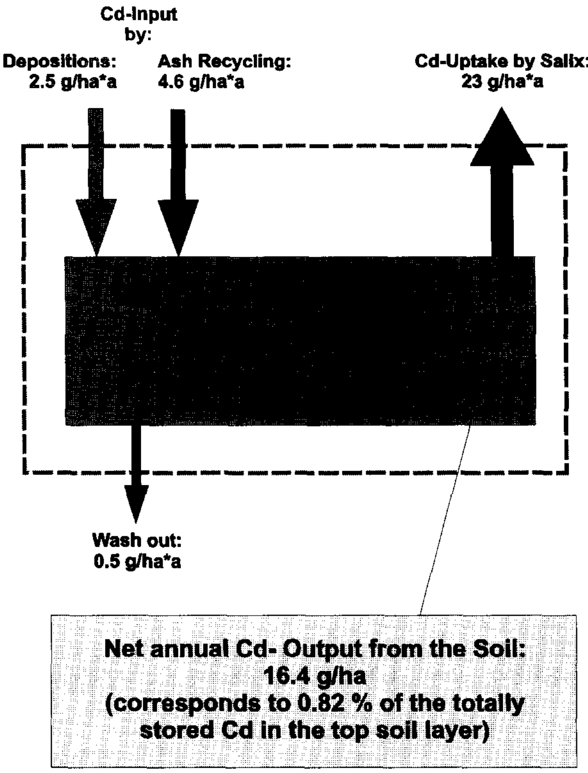

Fig. 3. Cadmium balance for an agricultural ecosystem under consideration of sustainable thermal biomass utilization and the technology of fractionated heavy metal separation in the combustion plant.
Explanations: Area used for short rotation forestry with Salix; annual harvested biomass: 10 t (dry basis) ha^{-1}; soil density (0–30 cm): 1300 kg m^{-3}; the biomass combustion plant where the Salix fuel is fired is equipped with new technology for fractionated heavy metal separation in the different ash fractions: the filter fly-ash fraction, containing 80% of the total cadmium input by the fuel is separated from the ash cycle, the rest of the ash is recycled to the soil.

sive Swedish research projects have shown that Salix removes cadmium from the soil and therefore serves as a first concentration step for this metal, taking up about 2.3 ppm (dry basis) on average ([11,12]). A technologically optimized biomass combustion system will then form a second concentration step, separating cadmium from the major part of the ash that may be recycled and upgrading it in the filter fly-ash fraction.

The cadmium balance for a normal Austrian agricultural soil that shows a moderate cadmium accumulation in the top soil layer (the cadmium concentration of non contaminated agricultural soils in Austria lies between 0.1 and 0.3 ppm (dry basis)) is calculated in Fig. 3. By establishing short rotation forestry with Salix on such an area and using the biomass produced as fuel in a new combustion plant that fulfils the requirements for a fractionated heavy metal separation in the different ash fractions, about 80% of the cadmium taken up by the biomass should be bound in the filter fly-ash fraction (according to results of test runs already achieved in a pilot plant ([13]).

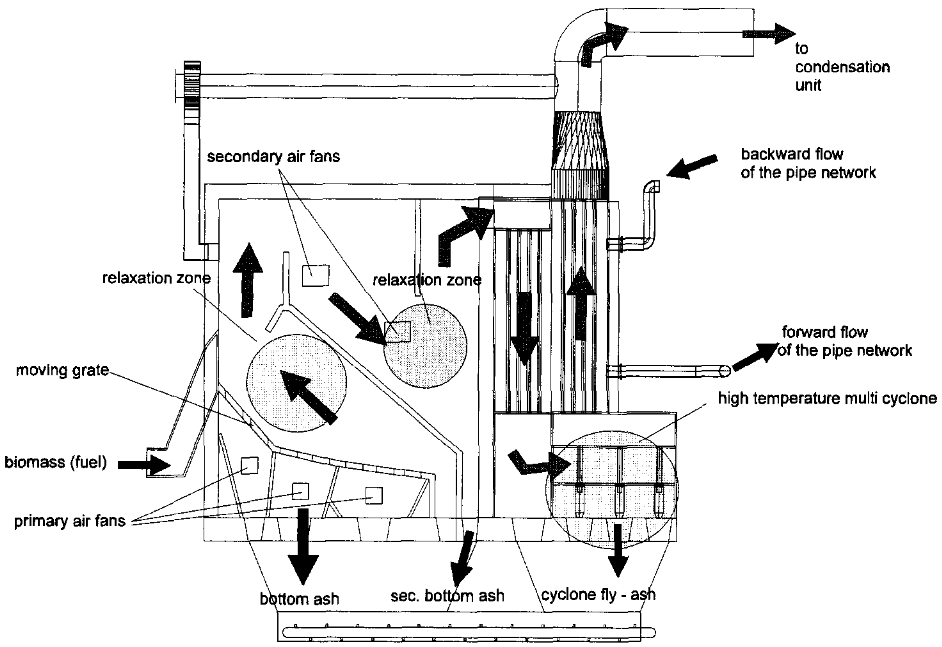

Fig. 4. Scheme of an innovative combustion technology with integrated fractionated heavy metal separation.

Consequently about 0.8–1.3% of the total amount of cadmium in the top soil layer could be removed yearly from the ground although about 90% of the total ash produced from Salix combustion would be recycled (a mixture of bottom ash and cyclone fly-ash). The large net cadmium removal from the soil results from separating the small but heavy metal reach filter fly-ash fraction from the ash cycle and could reduce the cadmium concentration in the soil in about 50 years to 0.3 ppm (40% net reduction).

Fig. 4 shows a sketch of a new combustion technology that would satisfy the requirements for a fractionated heavy metal separation. The idea is to precipitate as much fly-ash as possible at high temperatures (in the furnace or in high temperature cyclones) in order to keep volatile heavy metals out of this fraction. To achieve this the combustion chamber has to be redesigned. A relaxation zone for the flue gas over the grate combined with a reducing atmosphere in the primary combustion chamber ($\lambda < 1$) should guarantee that on the one hand the fly-ash production is as low as possible and that on the other hand the volatilization rate for metals is high. A second big relaxation zone for the flue gas in the secondary combustion chamber combined with a high temperature cyclone should enable recovery of a secondary bottom ash fraction as well as a cyclone fly-ash fraction precipitated at high temperatures. By means of this technology especially cadmium desublimation/condensation on fly-ash particles of these ash fractions should be prevented.

In this manner volatile heavy metals are kept out of the largest part of the ash. It is, however, necessary to recover them in the process in order to reduce their dissipation. This is realized by a highly efficient flue gas condensation unit, where the gas temperature is reduced to about 40°C. At this temperature, a very high fraction of cadmium can be captured and concentrated in the relatively small fraction of condensation sludge. First results from experiments show that this technology has the potential of concentrating 80% of the cadmium entering the biomass combustion plant in about 6% of the ash and to reduce the concentration of cadmium in the rest of the ash (mixture of bottom ash, secondary bottom ash and cyclone fly-ash) to about one third of the amount in blended ash from state-of-the-art biomass combustion plants that are equipped with conventional combustion technology.

6. Conclusion

Ash is a crucial aspect of the concept of sustainable, CO_2-neutral thermal biomass utilization. In contrast with the carbon cycle, mass fluxes of nutrients contained in the ashes may not be closed satisfactorily with state-of-the-art combustion technology. The main culprits for this are volatile heavy metals, especially cadmium, that pollute the fraction of the ash leaving the combustion system at the cyclone.

As a medium term solution, cyclone fly-ash and bottom ash may be mixed in the same ratio as they are produced in the plant. This results in a blended ash that might be recycled with caution in forests and on agricultural land.

A long-term solution, however, requires technological development. First the technology of combustion and ash management has to be changed in a way to concentrate cadmium and other volatile heavy metals in a small ash fraction and to recycle the overwhelming part of the ash (and the nutrients) to soil. Thus, an effective closing of material cycles and a sustainable energy provision from biomass is possible. Preliminary results show a high potential for cadmium retention in the small fraction of condensation sludge by technologies that are currently under development.

The application of such technologies will not only improve the ecological acceptability of biomass combustion systems. They might also establish thermal utilization of biomass as an effective step in technologies to reduce the cadmium load dissipated to the environment by other industrial processes.

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