



## Review article

# Fate and distribution of heavy metals during thermal processing of sewage sludge



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## ABSTRACT

Thermal processing of sewage sludge (SS) has received increasing attention in recent years. Thermal processes valorise the carbon rich organic fraction of SS, while effectively reducing SS volume. However, the fate and distribution of heavy metals (HMs) during thermal processing of SS is an important issue to address because it has impact on the generation of secondary pollutants and the environmental acceptability of the residues for reuse and reclamation. The refractory metals (thermally stable, i.e. Cr, Mn and Ni) are less volatile at typical temperature ranges (200–1100 °C) of thermal processes, and they are enriched in the residues. On the contrary, HMs with lower thermal stability (i.e. Hg, Cd, As and Pb) are prone to volatilisation. However, volatilisations and enrichments of HMs in the residues strongly depend on the characteristics of SS and nature of the thermal process. This review article discusses the volatilisation, enrichment and speciation (or stabilisation) of HMs in the residues formed during thermal processing of SS (incineration, pyrolysis, gasification and hydrothermal treatment). First, it summarises the fundamental aspect of SS in each thermal process. The influencing factors on the fate and distribution of HMs are discussed in terms of process principles, reactor types, operating conditions, pre-treatment of SS, use of additives and co-processing with secondary feedstocks. The use of advanced analytical techniques and modelling tools to analyse the complexity of HMs redistribution during thermal processing is described. Practical and economic challenges associated with HMs in SS during operation of full-scale thermal processing facilities are also addressed. Finally, a brief comparison of HMs redistribution and stabilisation during SS incineration, gasification, pyrolysis and hydrothermal treatment is provided.

## 1. Introduction

Population growth, economic development, rapid urbanisation, rising environmental and public health standards, and improved sanitary systems have resulted in a vast number of wastewater treatment plants (WWTPs) worldwide. Consequently, the volume of sewage sludge (SS) generation from WWTPs has been steadily increasing over the years. SS generation in Europe has been estimated to be around 12 million tonnes by 2020 [1]. Over 20 million tonnes of annual SS production has been reported in China [2,3]. Moreover, it has been reported that SS treatment counts more than 50% of the total operational cost of WWTPs [4]. Thus, development of sustainable strategy for SS management has become imperative. Advanced wastewater treatment technologies to minimise SS generation, recycling and recovery of

useful materials from SS, and energy extraction have received significant attention in recent decades [5].

SS is a heterogeneous mixture of microorganisms and undigested organic and inorganic components with high moisture content. The undigested organic matter contains proteins and peptides, lipids, polysaccharides, plant macromolecules alongside with micro-pollutants (i.e. polychlorinated biphenyls (PCBs), polycyclic hydrocarbons (PAHs), pesticides, surfactants, by-products of pharmaceutical and personal care products). Minerals such as quartz, calcite and traces of heavy metals (HMs) such as Cu, Zn, Cd, Pb, Ni, Cr, Hg and As represent the inorganic fraction of SS [6–9]. SS also contains considerable concentrations of N, P and K, which are valuable nutrients to plant growth. Thus in early days, agricultural use and landfilling were the common management strategies of SS. However, the existence of harmful

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### Nomenclature

AAS	atomic absorption spectrometer	MSW	municipal solid waste
AFS	atomic fluorescence spectrometer	NAA	neutron activation analysis
ANN	artificial neural network	OMC	operating and maintenance cost
BCR	community bureau of reference-European commission	PAH	polycyclic aromatic hydrocarbon
BET	Brunauer-Emmet-Teller	PCB	polychlorinated biphenyl
DTPA	diethylenetriaminopentaacetic acid	SEP	sequential extraction procedure
EDTA	ethylenediaminetetraacetic acid	SS	sewage sludge
EDX	energy dispersive X-ray spectroscopy	SSNMR	solid state nuclear magnetic resonance
ESP	electrostatic precipitator	TCLP	toxicity characteristic leaching procedure
FESEM	field-emission scanning electron microscopy	TEM	transmission electron microscopy
FTIR	Fourier-transform infrared spectroscopy	TGA	thermogravimetric analysis
GC-MS	gas chromatography-mass spectrometry	WtE	waste-to-energy
HM	heavy metal	WWTP	wastewater treatment plant
HPLC	high-performance liquid chromatography	XANES	X-ray absorption near edge spectroscopy
ICP-MS	inductively coupled plasma-mass spectrometry	XPS	X-ray photoelectron spectroscopy
ICP-OES	inductively coupled plasma-optical emission spectroscopy	XRD	X-ray powder diffraction
		XRF	X-ray fluorescence

elements and constituents (i.e. HMs, pathogenic bacteria, viruses, toxic organic compounds), which could pose acute and chronic toxicities to the environment and human health, have imposed stringent regulations to restrict the direct use of SS [5,10–13]. The organic fraction of SS comprises high content of carbon, providing calorific value (in its dry form 10–20 MJ kg<sup>-1</sup>) close to low grade coal (lignite) or biomass [14–16]. In this context, fuel generation (i.e. heat, hydrochar, bio-oil, syngas) from SS (sludge-to-energy, sludge-to-fuel technologies) via thermal processes (i.e. incineration, gasification, pyrolysis, hydrothermal) has received increasing attention, while destructing the toxic organic compounds and pathogens accompanied with effective volume reduction in short reaction time [4,7,16,17]. Moreover, thermal decomposition of SS in oxygen-limited environment (i.e. pyrolysis, hydrothermal) leads to carbon rich solid product, which has secondary applications [18–20]. Hence, modern practices have focused on thermal processing as a sustainable path of SS management. Fig. 1 shows the simplified process path adopted in conventional and modern thermal strategies of SS [21]. Table 1 summarises the advantages and disadvantages of SS reuse strategies and a comparison with various thermal processes [5,6,10,11,22–24]. Despite the promising prospects, conventional methods are still prevailing for SS management [16,25–27].

Incineration is the most common thermal process for SS treatment [25,26]. However, gasification, pyrolysis and hydrothermal processes have gained increasing interest owing to their advantages over

incineration (Table 1) [10,11,28–30]. Integrated thermal processes such as pyrolysis-gasification, pyrolysis-combustion, hydrothermal-pyrolysis as well as co-feeding of SS as a secondary fuel source during thermal processing of biomass and coal have also been explored for the effective management of SS [17,31–33]. The high moisture content and inorganic constituents of SS are the major constraints for thermal processing; for instance, removal of moisture consumes considerable amount of energy, while inorganic fraction causes ash related issues such as ash fusion and agglomeration, and volatilisation of HMs [34–36]. Some studies have shown that up to certain extent moisture and some inorganic constituents in SS could assist the thermal conversion/degradation of organic fractions [34,37–40]. Furthermore, owing to heterogeneous nature of SS, thermal processing products are heterogeneous or contaminated with pollutants such as HMs, PAHs and PCBs. Thus, further treatments may be needed prior to their use in secondary applications or even for ultimate disposal [41,42].

The presence of HMs in SS causes many challenges during thermal processing. The elevated temperatures used in thermal processes redistribute the HMs in SS by forming various physical and chemical phases. Table 2 shows the common forms of HM compounds and their properties. Emission of volatile metallic compounds begins once the temperature reaches the boiling point of the metal compounds. The volatile compounds start to condense or nucleate at lower temperatures in downstream units. Besides volatilisations, HMs (especially the refractory metals) are also redistributed by the entrainment process [43].

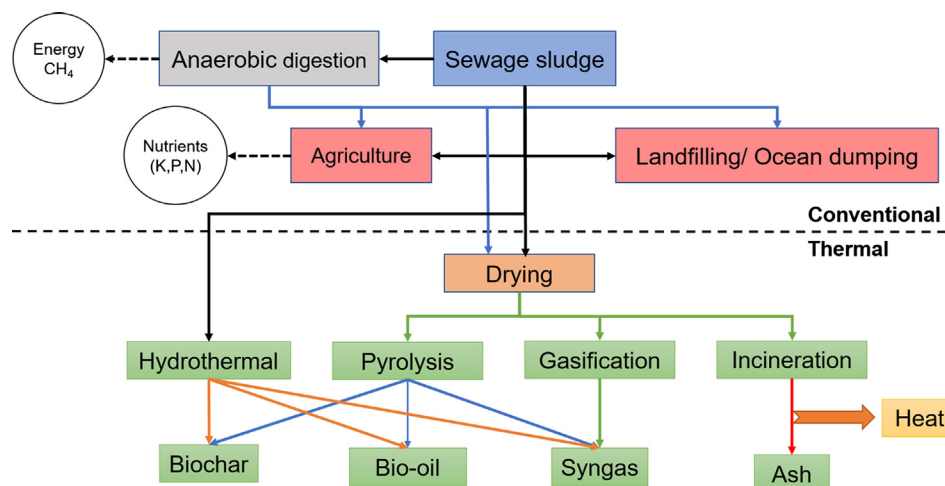


Fig. 1. Simplified process path of SS adopted in conventional and thermal processes.

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