Journal of Industrial and Engineering Chemistry xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Journal of Industrial and Engineering Chemistry

journal homepage: www.elsevier.com/locate/jiec

Survey of the mercury-containing wastes released from various sources in Korea

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ARTICLE INFO

Article history:
Received 11 October 2017
Received in revised form 4 December 2017
Accepted 13 December 2017
Available online xxx

Keywords:
Minamata convention
Basel convention
Fly ash
Bottom ash
Sludge

ABSTRACT

An extensive study of wastes containing mercury (Hg) released from carefully selected industrial sources in Korea was conducted. First, 89 sources associated with 21 Hg waste-producing industries were selected for investigation. The types of waste generated, the treatment methods used, the total annual amounts of waste released, and Hg concentrations of waste samples were then thoroughly identified. Two wastewater treatment sludge samples were classified as waste with high Hg concentrations (US EPA; >260 mg/kg). Based on the 156 waste samples analyzed in this study, the total annual amount of Hg released was estimated to be close to 33 t.

Introduction

To protect human health and the environment from the threat of mercury (Hg), the 21st Governing Council of the United Nations Environment Programme (UNEP GC-21) resolved to initiate a global Hg assessment in 2001 [1]. In 2013, to finalize the negotiations for a global legally binding instrument on Hg, the text was opened for signature at a Diplomatic Conference (Conference of Plenipotentiaries) that was held in Minamata, Japan [2]. "The Minamata Convention on Mercury" entered into force on August 16, 2017 [3].

The EU has enforced regulations that ban the export of certain Hg compounds and mixtures and control the safe storage of metallic Hg. These regulations, specifically Regulation (EC) No 1102/2008 (22 October 2008), took effect on March 15, 2011 [4]. Moreover, the U.S. enacted the Hg Export Ban Act as of January 1, 2013 to prohibit the export of metal Hg and certain Hg compounds [5]. The U.S. has retained more than 5600 t of Hg for raw materials since 1970 and undertaken the transfer of Hg to permanent storage facilities. Japan exports approximately 190 t of Hg derived from the recycling of Hg-containing products annually, with more than 70 t derived from nonferrous metal smelters [6].

The National Institute of Environmental Research in Korea has conducted several studies on Hg release from various sources [7–9]. However, attention must be given to Hg-containing wastes since considerable amounts are still released from various industrial facilities, even though the commercial use of Hg has recently decreased [10]. There is an urgent need for a database of Hg-containing wastes, which will lead to corresponding national standards as well as management policy.

In this study, a total of 156 samples collected from 89 carefully selected major industries that release Hg-containing wastes were analyzed for their Hg contents.

Experimental

Investigation methods

As shown in Fig. 1, a total of 89 facilities were selected from 21 Hg waste-producing industries that had been identified by UNEP and the Basel Convention using the following selection criteria: (1) domestic operation of the facilities, (2) the existence of preceding studies, (3) the amount of waste released, (4) the estimated amount of Hg in the waste, and (5) regional facilities.

To investigate the state of Hg-containing waste release, official documents issued by the Ministry of Environment and survey questionnaires were sent to the selected companies to obtain their full cooperation. The results were used to investigate the types and amounts of the raw materials used, the types of final waste produced, the annual amount of waste released and the treatment methods and technologies applied to the final wastes. Two samples

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https://doi.org/10.1016/j.jiec.2017.12.026
1226-086X/

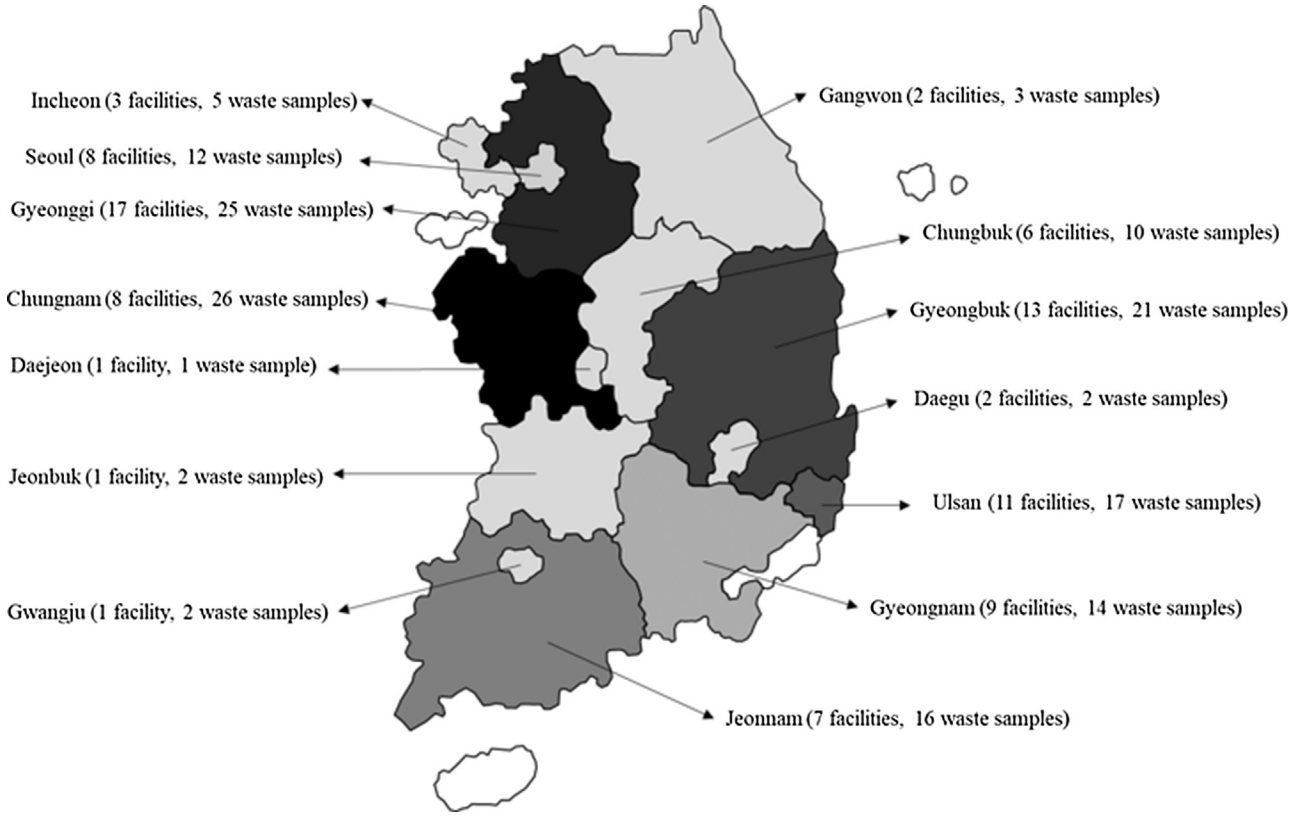

Fig. 1. A distribution map of sample collection sources.

were collected from each source (three from energy-related facilities) at an interval of more than one day (half a day for waste/wastewater treatment facilities).

Analysis methods

The Hg concentrations of the collected samples were measured using two standardized testing methods, US EPA methods 7471B and 7473 [11,12]. In addition, the moisture concentrations of the samples were measured using the waste treatment testing method [13].

The moisture concentration was measured as follows. A beaker was dried at 110 °C for one hour, and the dried beaker was inserted into a desiccator to be cooled. A sample was thinly spread on the floor of the beaker, and its weight was measured. The beaker containing the sample was dried at 110 °C for four hours and cooled in a desiccator, and its weight was measured. The moisture content was then calculated as

$$\text{Moisture content}\,(\%) = \frac{W_2 - W_3}{W_2 - W_1} \times 100$$

where W_1 indicates the weight of the beaker after it was dried at 110 °C for one hour and cooled in the desiccator, W_2 indicates the weight of the beaker containing the sample, and W_3 indicates the weight of the beaker containing the sample after it was dried at 110 °C for four hours and cooled in the desiccator.

US EPA method 7471B is a method for measuring the total amount of Hg in a sample of soil, grit, sediment, or wastewater treatment sewage sludge. The samples were preprocessed with acid, such as nitric acid, hydrochloric acid, sulfuric acid or potassium permanganate. The Hg in the oxidized samples was reduced and vaporized using a Sn(II) solution ($SnCl_2$) and measured using an RA-915$^+$/RP-91 CVAAS-type Hg analyzer (Lumex Ltd., St. Petersburg, Russia). The detection limit of the Hg analyzer was 0.5 ng/L, and the measurement range was 100–10,000 ng/L.

US EPA method 7473 enables the direct analysis of a solid or liquid sample using a pyrolyzer-type Hg analyzer (DMA-80, Milestone Srl., Bergamo, Italy) without preprocessing. The samples were initially dried and then thermally decomposed in a continuous flow of oxygen. Combustion products were carried off and further decomposed in a hot catalyst bed. Hg vapors were trapped on a gold amalgamator and subsequently desorbed for quantization. The Hg content was determined using atomic absorption spectrophotometry at 254 nm. The detection limit was 0.005 ng, and the range of measurement was 1000 ng or less. The maximum Hg concentration that can be measured is 30,000 ng/g when samples with a mass of 0.03 g are used. Samples with Hg concentrations that exceed 30,000 ng/g cannot be measured using the DMA-80 analyzer. Therefore, in this study, the Hg concentrations of the waste samples were first determined using RA-915$^+$/RP-91, and cross-analyses were performed on the samples that were measurable using the DMA-80 analyzer.

Results and discussion

The selected Hg waste-producing industries and the treatment methods used

The release sources of domestic Hg-containing wastes and the types of wastes are listed in Table 1, with reference to the list of Hg-containing wastes per industry issued by UNEP in 2011 and the list of domestic Hg distribution and release sources issued by the Korean Ministry of Environment in 2009 [14,15]. The sample

Please cite this article in press as: S. Kim, et al., Survey of the mercury-containing wastes released from various sources in Korea, J. Ind. Eng. Chem. (2017), https://doi.org/10.1016/j.jiec.2017.12.026