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Substance Flow Analysis of Mercury from Industrial and Municipal Wastewater Treatment Facilities

Hyunhee Kim[a,1] , Yong-Chul Jang [a,*,2] and Yong-Seok Hong[b, 3]

*[a] * Department of Environmental Engineering, Chungnam National University, 99 Daehac-ro, Yuseong-gu, Daejeon, 34134, South Korea.*
[b] Department of Environmental Engineering, Daegu University, Daegudae-ro 201, JinRyong, Kyoungsan, Kyoungsangbuk-do, 38453, South Korea.

[1]Orcid: 0000-0002-0488-315X, [2]Orcid: 0000-0001-5435-2915, [3]Orcid: 0000-0001-5944-6515

Abstract

Mercury has become a persistent pollutant of major concern in international community as UNEP (United Nations Environment Programme) has recently adopted Minamata Convention on Mercury. In this study, substance flow analysis (SFA) of mercury was performed on waste materials from two industrial wastewater treatment facilities. A number of waste and wastewater samples from the facilities were collected to determine the levels and flow of mercury. Other relevant data such as sludge generation, chemical addition, and wastewater treatment rates were obtained through the field investigations, literature review from available reports and statistics. Data reconciliation with SFA Software (STAN) was used for mercury flow at the sites by decreasing data error of this study by adopting Least Square Method and Error Propagation Theory. Based on the results of SFA, it was found that major flow of mercury at wastewater treatment facilities is present in sludge during the treatment processes. As a result of this study, it was found that mercury flow from industrial wastewater treatment facility was estimated to be 12.45 g/day (equivalent to 4.5 kg/yr), mainly from sludge treatment process from the industrial wastewater treatment facility. In case of municipal wastewater treatment facility, 36.3g/day of mercury (equivalent to 13.3kg/yr) is emitted to sludge during municipal wastewater treatment process. More detailed examination on more diverse wastewater treatment facilities is still needed to eliminate uncertainty of mercury flow from the sites.

Keywords: Mercury, SFA (Substance Flow Analysis), wastewater treatment, sludge, STAN.

INTRODUCTION

Mercury has become a chemical of major concern in international community as recently UNEP (United Nations Environment Programme) has adopted Minamata Convention on Mercury as an international action all over the world [1]. Mercury as a substance having harmful effects on human health and environment with long distance movement and bio-accmulation is globally managed as one of the priority management target substances. In order to properly manage the chemical in the environment, the sources and behavior, and quantitative flow of mercury have to be determined. Several material and substance flow of mercury have been carried out [2-7]. However, there are still needed for quantitative evaluation, behavior and flow of mercury in many industrial sources.

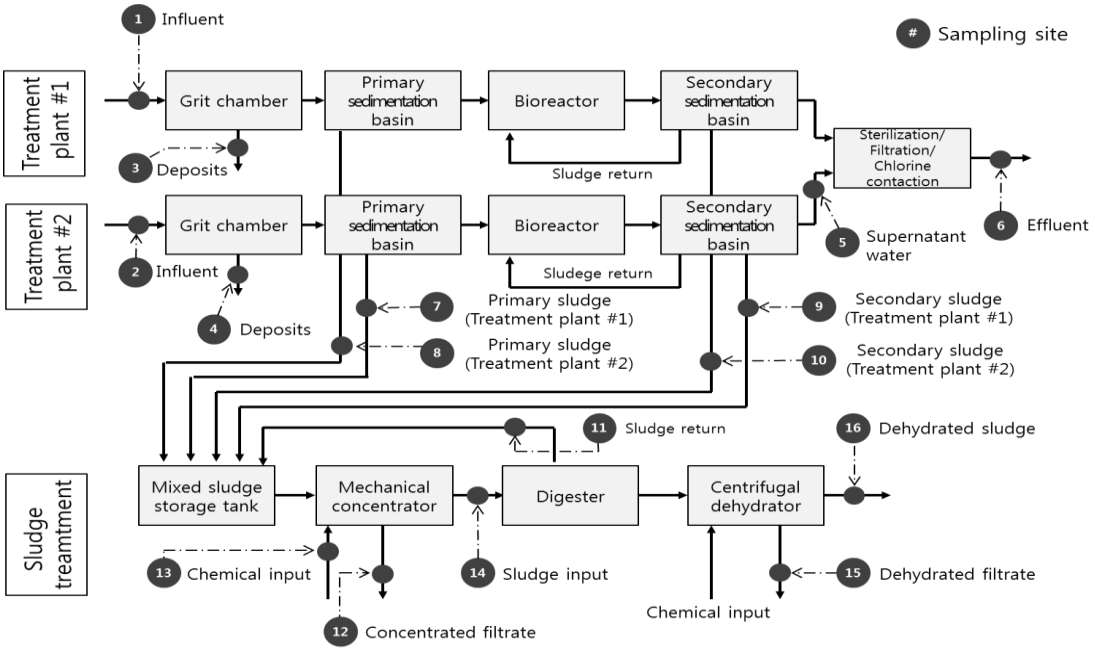
In this paper, substance flow analysis (SFA) of mercury was performed on waste materials from both industrial wastewater treatment facility and municipal wastewater treatment facility by analyzing mercury from various sources from the facilities. Data regarding the emission, flow and distribution of mercury and its waste from various industrial processes and sources are very limited so far. It is important to quantitatively determine mercury flow in the processes and sources for sustainable management and phase-out of the toxic material. Thus, the objective of this research is to determine the behavior and quantitative flow of mercury in wastewater treatment system to be supported by Material flow analysis (MFA) software STAN.

MATERIALS AND METHOD

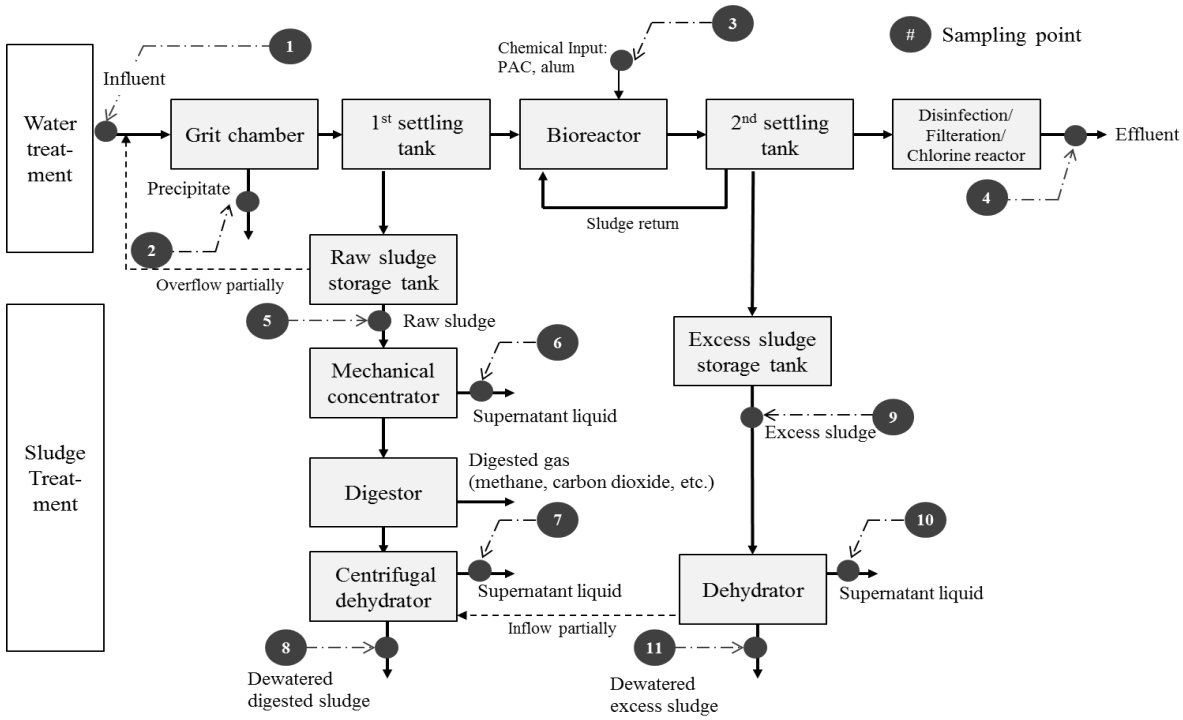
Sampling and Wastewater Treatment Processes

The sample sites were selected for mercury flow at industrial wastewater treatment facility and municipal wastewater treatment facility in 2014. The sites are located at Kyoungbuk Province and Daejeon Metropolitan City in South Korea, respectively. The industrial wastewater treatment facility consists of two treatment locations (Site #1-57,000 ton/day, Site #2 43,000 ton/day) with secondary treatment process. The main wastewater treatment processes include grit chamber, primary settlement tank, biological treatment (bioreactor), secondary settlement tank, sludge concentrator, sludge digester, filtration and dehydration process. On average, 1,200 ton of sludge is generated during the wastewater

treatment facility. The detailed industrial waste water treatment process can be found in Figure 1(A). In case of municipal wastewater treatment facility, the treatment process is similar to that of the industrial wastewater treatment facility, but it is a large scale of treatment. The capacity of municipal wastewater treatment is 600,000m^3/day. The actual treatment rate reaches 390,000 m^3/day. The municipal sludge is generated at 7,500 ton/day. The digested sludge and excess hydrated sludge are found to be 31.3 and 116 ton/day, respectively. The detailed wastewater treatment process is found in Figure 1(B).



(A) Industrial wastewater



(B) Municipal wastewater

Figure 1. Mercury sampling points of the wastewater treatment process at industrial wastewater treatment facility(A) and municipal sewage treatment facility(B)

Sample Collection and Analysis

A total of 45 samples for mercury from 15 sampling locations with three consecutive days were collected from the industrial wastewater treatment facility, while 24 samples were collected from the municipal wastewater treatment facility. Most samples include influent, effluent, raw sludge, dewatered sludge, dehydrated filtrate, raw chemical as coagulants. The repeated samples of mercury were averaged to employ its concentration for SFA. Figure 1 displays the detailed locations of the samples for mercury analysis from the two treatment facilities.

A stainless steel scoop was used to remove the top 15 cm of solid sludge to expose the sampling area. The composite sludge sample in the bowl was mixed and then transferred into appropriate containers. Approximately 50-g of sludge sample was weighed into a plastic container in a cooler. Aqueous samples (e.g., influent, effluent, raw alum chemicals) for mercury analysis were directly collected from inlet and outlet of the facility in 2-liter glass jars with Teflon-lined lids. Both aqueous samples and solid samples were analyzed by combustion and CV AAS (DMA-80 Analyzer). The detailed analytical method is described in US EPA SW-846 7473 [8]. The detection limit of total mercury is 0.1 ng/g.

Substance Flow Analysis

Substance flow was analyzed based on data from field investigation and waste sampling. Such flow analysis was accomplished using MFA Stan 2.5 Software [9]. The system boundary of substance flow analysis was set to the wastewater treatment process in the facility including sludge treatment processes. In order to reduce data error, this study utilized data reconciliation of Stan 2.5. Data revision of Stan 2.5 was accomplished based on least square method (Eq. 1) and error propagation theory (Eq. 2), as shown below in equation [10].

$$\chi^2 = \min\left(\sum_{i=1}^{N}\left(\frac{y_i - y^*}{\sigma_i}\right)\right) \quad \text{Eq. (1)}$$

$$\sigma_y^2 \approx \left(\frac{\partial Y}{\partial X_1}\right)^2_{\overline{X_1}\,\overline{X_2}} \cdot \sigma_{X_1}^2 + \left(\frac{\partial Y}{\partial X_2}\right)^2_{\overline{X_1}\,\overline{X_2}} \cdot \sigma_{X_2}^2 + \cdots \quad \text{Eq. (2)}$$

RESULTS AND DISCUSSION

Mercury analysis at wastewater treatment facility

Mercury flow was determined by a number of samples collected from the industrial wastewater treatment facility. **Table 1** shows the results of mercury flow of various sources such as influent, effluent, sludge, concentration filtrate, dehydrated sludge and filtrate. It should be note that the input and output analysis has been conducted to maintain mass balance of mercury in the defined boundary system. Such analysis was used for mercury concentration and flux in the wastewater treatment process at the site. Large fraction of mercury flow is present in sludge during the wastewater treatment process.

Table 1: Results of mercury analysis from samples collected from industrial wastewater facility

Sampling Points	Description	Results ranges[1)] (g/day)		Mass balance result (g/day)	
		Min	Max	Average	Std
1	Plant#1 influent	8.40	24.43		
2	Plant 2_influent	5.95	12.81	Not included	
3,4	Plant 1,2_deposits	0.08	0.12	in system boundary	
6	Effluent	0.05	45.93		
7	Plant 1_raw sludge	0.12	34.73	11.16	5.07
8	Plant 2_raw sludge	0.11	0.37	0.15	0.12
9	Plant 1_sludge	0.10	1.66	0.68	0.62
10	Plant 2_sludge	0.07	1.20	0.39	0.30
12	Concentrated filtrate	0.00	2.02	1.15	0.69
13	Chemical input	0.02	0.12	0.07	0.04
14	Sludge input	0.41	15.07	11.29	5.01
15	Dehydrated filtrate	0.20	0.29	0.14	0.03
16	Dehydrated sludge	19.60	59.10	11.15	5.01
Total input/output (sludge)				1.0	

1) Results range refers minimum and maximum amounts of mercury input and generation using mercury sampling results and flux.

In the municipal wastewater treatment facility, the flow rates and mercury analytical results are shown in **Table 2.** The highest mercury was found in digested dehydrated sludge (0.374 mg/kg). Based on the flow rates and mercury content, mercury flow from the facility was determined, as shown in **Table 3**. The major flow of mercury at the facility ranged from 12.12 g/day from raw sludge to 24.1 g/day from excess sludge.

Table 2: Results of input and output from municipal wastewater plant

Sampling Points	Description	Flow rate (ton/day)		Mercury results	(mg/L or mg/kg)
		Average	Std	Average	Std
1	influent	392,925	19,646	0.001	0.001
2	Settlement[1)]	1,078	53.9	0.025	0.008
3	Chemical input	12	0.6	0.083	0.007
4	Effluent	356,060	14,405	0.001	0.000
5	raw sludge	3,412	881	0.004	0.003
6	Concentrated filtrate from filter press	2,460	126	0.000	0.000
7	Concentrated filtrate from digestion	N/A	N/A	0.001	0.002
8	Digested dehydrated sludge	31	5.3	0.374	0.081
9	Excess sludge	4,039	268	0.111	0.111
10	Concentrated filtrate from excess sludge	N/A	N/A	0.001	0.001
11	Excess hydrated sludge	116	3.4	0.182	0.034

1) Unit: ton/year

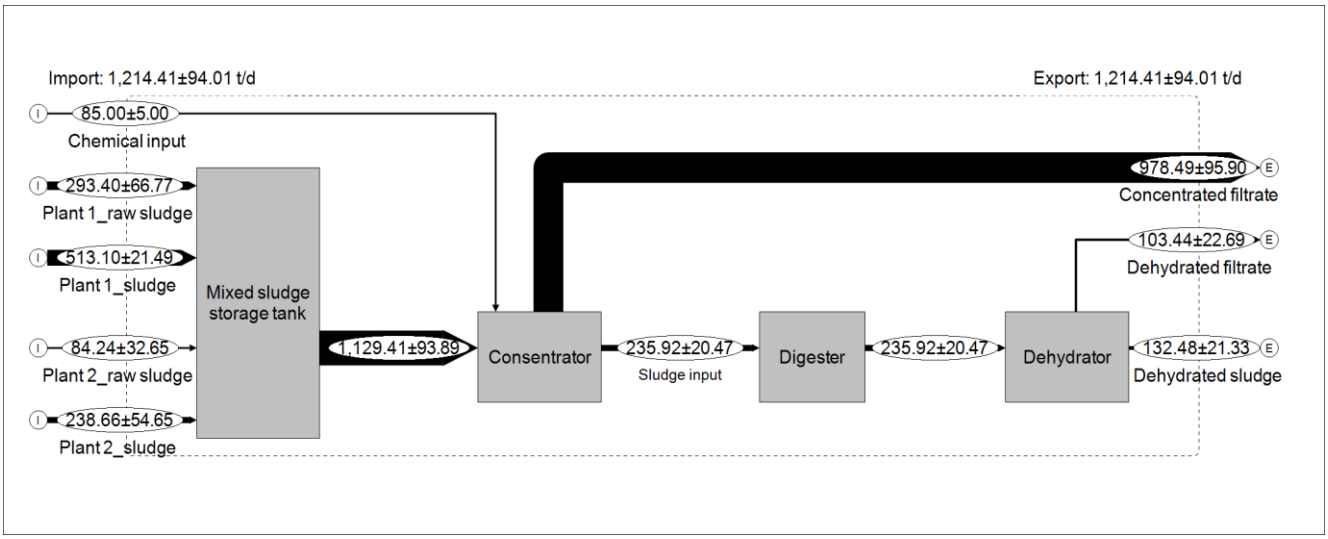
Table 3: Results of mercury analysis from samples collected from municipal wastewater facility

Sampling Points	Description	Mercury flow[1)] (g/day)	Mass balance result (g/day)	
			Average	Std
1	influent	235.8	Not included in system boundary	
2	Settled deposits	0.07		
3	Chemical(alum)	0.97		
4	Effluent	231.44		
5	raw sludge	12.28	12.12	7.65
6	Concentrated filtrate from filter press	0.08	0.02	0.02
7	Concentrated filtrate from digestion	1.13	1.15	1.62
8	Digested dehydrated sludge	11.66	11.62	2.48
9	Excess sludge	389.9	24.20	9.42
10	Concentrated filtrate from excess sludge	3.73	2.41	3.53
11	Excess hydrated sludge	21.12	21.12	3.03
Total input/output (sludge)			1.0	

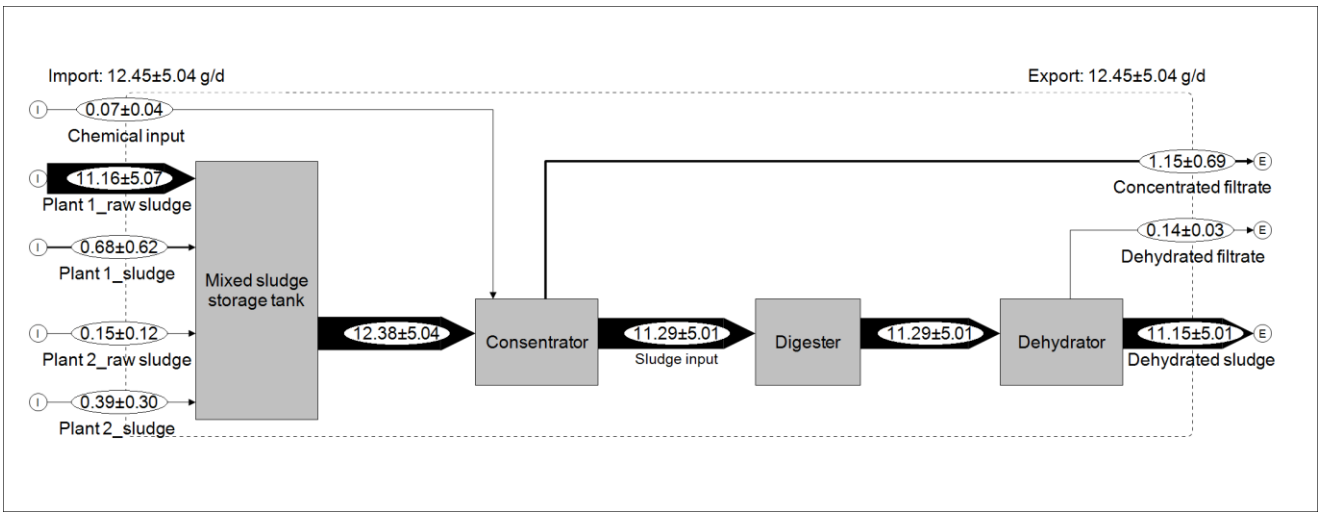
[1)] Mercury flow was determined by flow rate multiplied by mercury concentration.

Mercury flow at industrial and municipal wastewater treatment facilities

Figure 2(A) shows the flow of sludge at the industrial wastewater treatment processes. After the concentrator, most aqueous products move into concentrated filtrate (978 ton/d), while after the digestion, a large amount of sludge is generated as dehydrated sludge for further final treatment. **Figure 2(B)** shows SFA of mercury of the facility. As results of substance flow analysis of mercury in the wastewater treatment facility, it was analyzed that 1,214 tons of sludge was treated in a day. Mercury contained in sludge was determined to be 12.38g /day. It was found that most of mercury was discharged in dehydrated sludge, whereas most of sludge is recirculated into concentrated filtrate. It implied that mercury flowing into sludge treatment process in the facility was concentrated on raw sludge in the treatment plant. After sludge treatment process, mercury flow in the dehydrated sludge (approximately 132 ton/day) as final disposal was estimated to be 11.15g/day.



(A) Sludge



(B) Mercury

Figure 2: Flow of sewage(A) and mercury(B) at industrial wastewater treatment facility

Figure 3 illustrates mercury flow at the municipal wastewater treatment process. The mercury flow from raw sludge was found to be 12.79g/day. The higher mercury input is generated from excess sludge (24.1 g/day). After the sludge digestion, 11.62g of mercury is emitted in digested sludge, while dehydrated sludge contained 21.12 g of mercury per day.

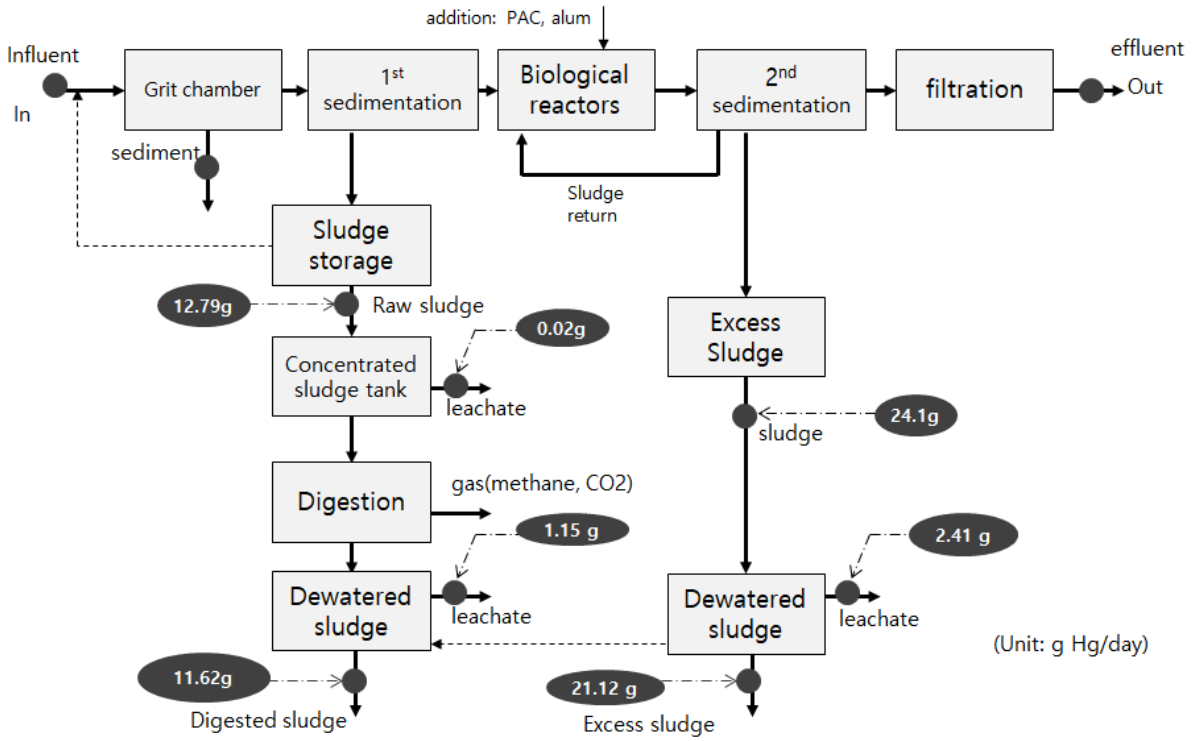


Figure 3: SFA of mercury in municipal wastewater treatment facility

Comparison study of mercury flow at wastewater treatment facilities

Table 4 presents a comparison of mercury flow at wastewater treatment facilities. Mercury in sludge ranged from 118 g/day to 12.45 g/day. The different values of mercury flow at wastewater treatment facilities may depend upon a number of factors including treatment flow rate and capacity, type of treatment process and influent, and sludge treatment process. Mercury content in wastewater (g/100,000m^3) at wastewater treatment facilities ranged from 9.2 g/100,000m^3 to 18.1 g/100,000m^3.

Table 4: Mercury in sludge and wastewater from industrial and municipal wastewater treatment facility

Wastewater treatment facility	Influent wastewater (m^3/day)	Mercury in sludge (g/day)	Mercury in wastewater (g/wastewater100,000m^3)
Industrial wastewater treatment facility	89,169	12.45	14.0
Municipal wastewater treatment facility	392.925	36.3	9.2
Balogh and Nollet (2008) [5]	651,456	118	18.1

CONCLUSION

In this study, SFA of mercury in industrial and municipal wastewater treatment facilitis is achieved by using Material Flow Stan 2.5 software. The result of SFA of mercury from the wastewater treatment facilities was found that the most amount of mercury was discharged as the digested dewatered sludge and concentrated sludge. As a result of SFA, a considerable amount of mercury can be mobilized by solid waste flow. Since mercury is present in primarily digested dewatered sludge and dewatered excess sludge, there may be required for appropriate measures and attention to manage the chemical in sludge treatment after wastewater treatment as a result of byproduct. Mercury flow analysis for diverse wastewater treatment facilities is still needed to representatively determine quantitative flow and behavior (emission, sinks) of mercury.

ACKNOWLEDGEMENTS

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