

Analysis of sewage sludge and cover soil by neutron activation analysis

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The Korean government reported that in 2005, 4395 tons/day of sewage sludge were generated from sewage disposal facilities in Korea and only 11.03% of it was reused. In addition, as a direct landfill of sewage sludge was forbidden from June 2003, research for a relevant disposal technique has been increasing. In this study, the aims were to analyze the collected sewage sludge samples and to evaluate the possibility for their reuse by a comparison of the elemental contents from a sewage sludge and a cover soil. Sludge samples were collected from a sewage disposal plant in Daejeon city and the cover soil was produced by a dilution of a sewage sludge with quicklime. Instrumental neutron activation analysis was employed to determine the elemental contents in the samples. Twenty seven elements were analyzed and their concentrations were compared.

Introduction

The Ministry of Environment in Korea reported that 294 sewage disposal facilities were being operated and 1,604,454 tons of sewage sludge was generated from them in 2005. For its disposal, 70.1% of it was dumped in the sea, 12.7% was incinerated, 6.2% was treated as landfill and only 11.0 % was reused.¹ Following a ban of ocean dumping by an international agreement, the Korean government has been making an effort to expand the reuse of sewage sludge. Inherently, sewage sludge has a high concentration of organic matter which can provide beneficial impacts on the soil. However, it may contain high contents of hazardous metals which can be toxic to the soil. Sewage sludge can be reused as compost, cover soil for landfill site, raw material for cement, etc., and the related researches have been carried out worldwide.^{2–4}

In this study, the possibility of the reuse of a sewage sludge was evaluated for a cover soil from the viewpoints of its elemental composition and content. For these objectives, sewage sludge samples were collected for a certain time period and the elemental content in the samples were determined by instrumental neutron activation analysis (INAA).^{5,6} Finally, the elemental concentrations of the sewage samples were compared with those of the cover soil.

Experimental

Sampling and sample preparation

Approximately 1 kg of a sewage sludge sample was collected monthly from a sewage disposal plant, which is located in Daejeon city in Korea, from June 2005 to January 2006. The collected samples were naturally dried and pulverized using an agate mortar (12–950C, Fisher). Finally, analytical samples were prepared by re-drying in an oven for 2 hours at 100 °C. In order to

make a cover soil sample, quicklime was purchased from a domestic mineral production company and sewage sludge samples collected for seven months were mingled with quicklime which was about 30% of the total sample weight in wet condition. The procedure for the preparation of a cover soil sample was the same as that for the sewage sludge. For neutron activation analysis, the prepared samples were divided into two target samples for detection of the short- and long-lived nuclides and they were put in polyethylene vials which were pre-cleaned using 0.1M nitric acid. The weights of the samples for the short-lived and long-lived nuclides were about 40 mg and 100 mg, respectively.

Neutron activation analysis

The NAA #3 irradiation hole with a pneumatic transfer system (PTS) was used for the activation of the prepared samples.⁷ The mean value of the thermal neutron flux measured at the NAA #3 hole was around $1.25 \cdot 10^{14} \text{ cm}^{-2} \cdot \text{s}^{-1}$ and the maximum irradiation time was limited to 1 minute, because of a rapid temperature increment during the sample irradiation. For the analysis using short-lived and long-lived nuclides, samples were irradiated for 2 seconds and 1 minute, respectively. Al-01%Au monitors were co-irradiated with the samples to correct the neutron flux variation due to the short irradiation time. For the measurement of the gamma-rays of the interesting nuclides, a HPGe detector (EG & G ORTEC, 25% relative efficiency, FWHM 1.85 keV at 1332 keV of ^{60}Co) coupled to a 16K-multichannel analyzer was used. During the gamma-ray measurement, the dead-time was kept below 10%. For a quality control, NIST SRM 2782 Industrial Sludge was chosen and analyzed under the same conditions as the samples. Elemental concentrations in the samples were determined by a software program available in the Windows environment, so called POWER NAA, which was developed for a fast and convenient calculation for the NAA.^{8,9}

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Results and discussion

Analytical results of the sewage sludge samples

Twenty five elements from the NIST SRM 2782 Industrial Sludge were analyzed and the results are shown in Fig. 1. The relative errors (%) of most of the determined elements were within 10% except for Au, Eu, Hf, Sb, and U which are reference values.

Twenty-seven elements in the sewage sludge samples were determined and the results are shown in Table 1. The concentrations of the major elements were Al 3.37%, Fe 0.74%, K 0.68%, Mg 0.60%, Na 0.22% and the relative standard deviations of these elements for seven months were 18.4%, 4.5%, 8.2%, 31.3% and 18.7%, respectively. Concentrations of the hazardous elements such as As, Cr, Cu, Mn, Sb and Zn, were $6.59 \pm 0.57 \text{ mg}\cdot\text{kg}^{-1}$, $41.8 \pm 0.86 \text{ mg}\cdot\text{kg}^{-1}$, $482 \pm 170 \text{ mg}\cdot\text{kg}^{-1}$, $557 \pm 113 \text{ mg}\cdot\text{kg}^{-1}$, $3.90 \pm 0.22 \text{ mg}\cdot\text{kg}^{-1}$ and $513 \pm 28 \text{ mg}\cdot\text{kg}^{-1}$, respectively. In accordance with the relative standard deviations which were less than 30%, it was found that the monthly variation of the elemental contents was not so large except for Mg, Cu, Ti and V.

Comparison with a cover soil

Thirty three elements in the cover soil sample could be determined by neutron activation analysis and the results are shown in Table 2. The concentrations of the major elements such as Al, Ca, Fe, K, Mg and Na were 1.86%, 15.1%, 0.81%, 0.67%, 0.46% and 0.090%, respectively. Among the thirty three elements, elemental contents for twenty seven elements determined both in the sewage sludge and cover soil samples could be compared in Fig. 2. Most of the elements in the cover soil revealed lower contents than those in the sewage sludge. In particular, the elements of Sr, Cu, Zn, Mn, Cl and Ba in the range of 100–1000 $\text{mg}\cdot\text{kg}^{-1}$ were considerably reduced. On the contrary, the concentration of six elements such as Ce, Cr, Hf, Fe, Sc and Th were similar or increased. Thus, it can be expected that the concentrations of some elements in the quicklime which was used for dilution, were higher than those in the sewage sludge. The reduction ratios for the hazardous elements such as As, Cr, Cu, Mn, Sb and Zn were evaluated as 0.93, 1.02, 0.10, 0.34, 0.51, and 0.40, respectively. From these results, it can be concluded that the analysis with quicklime sample, especially for As and Cr, should be accompanied with sewage sludge.

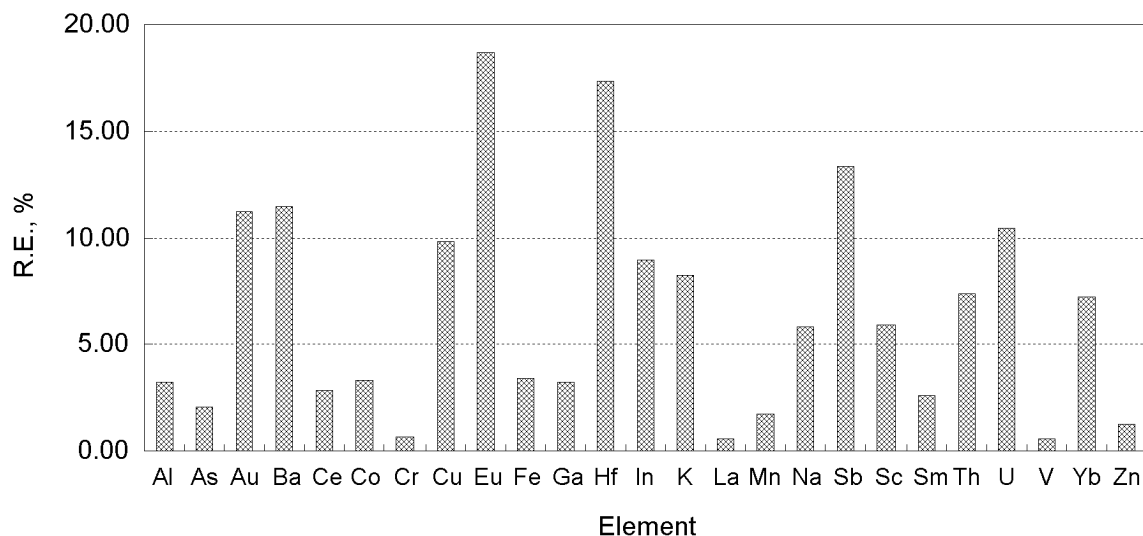


Fig. 1. Analytical results of NIST SRM 2782 Industrial Sludge by NAA

Table 1. Analytical results of sewage sludge samples collected during seven months (in mg·kg⁻¹)

Element	Range	Average	SD	Rsd, %
Al (%)	2.65~4.56	3.37	0.62	18.4
Fe (%)	0.69~0.81	0.74	0.03	4.5
K (%)	0.59~0.77	0.68	0.06	8.2
Mg (%)	0.37~0.83	0.60	0.19	31.3
Na (%)	0.15~0.27	0.22	0.04	18.7
As	5.78~7.57	6.59	0.57	8.7
Au	1.19~2.06	1.56	0.39	24.9
Ba	553~935	744	129	17.3
Br	25.3~29.4	26.9	1.3	4.9
Ce	14.5~20.9	17.9	1.8	10.0
Cl	568~936	692	152	22.0
Co	2.98~3.97	3.37	0.33	9.7
Cr	40.4~43.0	41.8	0.9	2.1
Cu	355~811	482	170	35.4
Hf	0.58~0.99	0.75	0.12	16.6
I	25.8~49.1	36.3	6.5	18.0
In	0.75~1.24	0.98	0.16	16.0
La	15.5~17.5	16.2	0.7	4.6
Mn	360~716	557	113	20.2
Sb	3.61~4.23	3.90	0.22	5.6
Sc	1.13~1.23	1.17	0.04	3.2
Sm	2.76~2.96	2.86	0.07	2.3
Sr	111~308	249	67	26.8
Th	1.82~2.51	2.08	0.24	11.5
Ti	399~1541	1019	388	38.1
V	11.1~33.0	21.3	7.1	33.2
Zn	453~543	513	28	5.4

Table 2. Analytical results of the cover soil samples ($n=3$) (in mg·kg⁻¹)

Element	Mean	SD
Al (%)	1.86	0.11
Ca (%)	15.1	0.5
Fe (%)	0.805	0.012
K (%)	0.67	0.06
Mg (%)	0.456	0.043
Na (%)	0.0897	0.0025
As	6.13	0.21
Au	0.19	0.02
Ba	210	11
Br	3.13	0.34
Ce	25.9	0.6
Cl	114	28
Co	2.83	0.12
Cr	42.8	2.4
Cs	1.23	0.17
Cu	49.9	18.3
Dy	1.39	0.16
Eu	0.40	0.03
Hf	2.06	0.35
I	8.48	2.01
In	0.167	0.021
La	13.9	0.5
Mn	189	8
Rb	37.0	3.0
Sb	1.99	0.18
Sc	1.61	0.04
Sm	2.05	0.02
Sr	163	12
Th	5.21	0.25
Ti	834	22
V	13.4	1.8
Yb	0.59	0.07
Zn	204.8	9.0

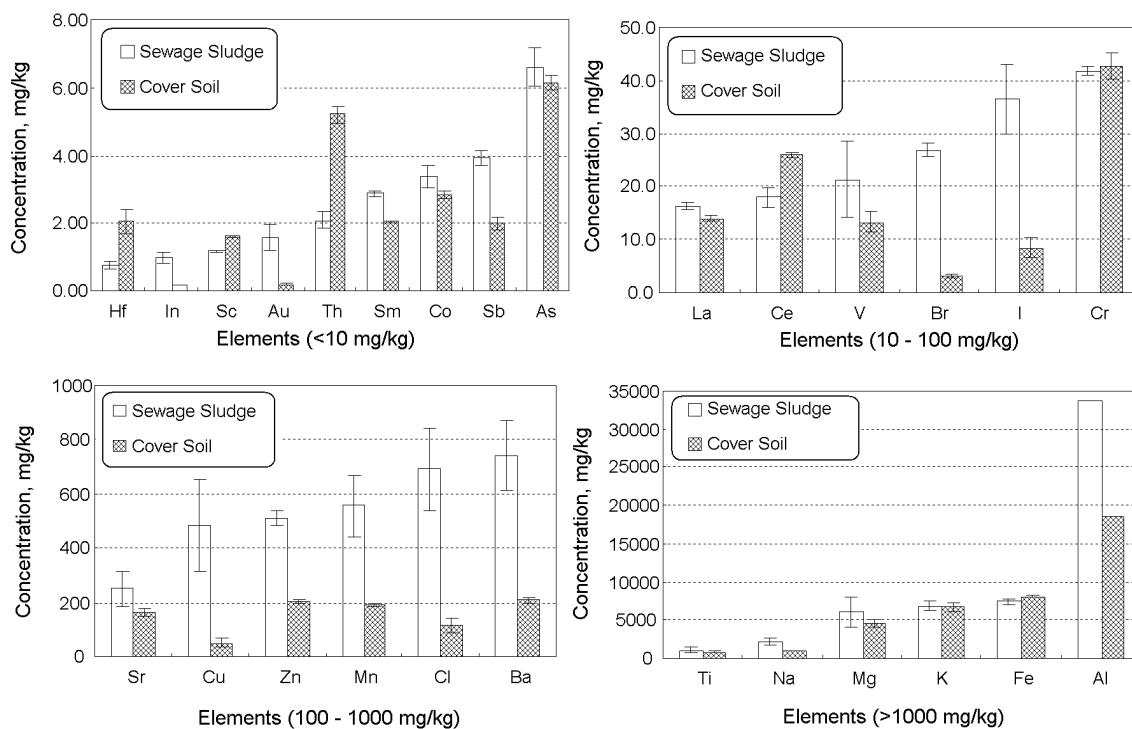


Fig. 2. Comparison of elemental contents between sewage sludge and cover soil samples

Conclusions

In this study, sewage sludge known as a waste material were analyzed by NAA and the possibility for its reuse was investigated by a comparison of the concentration of twenty seven elements with a cover soil. NAA has been effectively applied for the analysis of the environmental samples. However, we were not able to provide essential information on some toxic elements such as Pb, Ni and Cd. In order to maintain and enhance the applicability of NAA, collaborative works with other techniques are necessary.

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