Impact of waste co-combustion on mercury emission from cement production in Japan

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1. Introduction
During the last decade, anthropogenic mercury emission to the atmosphere has attracted great attention owing to global mobility of gaseous mercury and great toxicity to the ecosystem. Pacyna et al. (2002; 2006)¹² estimated the global atmospheric emission of mercury from anthropogenic sources to be 1912.8 Mg in 1995 and 2190 Mg in 2000, respectively. Thermal processes like coal-fired power plants, iron production, nonferrous metal smelting, medical waste incineration, and municipal and industrial waste combustions are major sources of mercury atmospheric emissions. In Japan, mercury atmospheric emission for 2002 were reported to be 27 Mg for year 2002 (Kida et al (2008))³. In Korea, it was evaluated to be 12.8 Mg for year 2007 (Kim et al. (2010))⁴. Mercury emission from cement production seems to be one of significant sources in Japan and Korea according to its large consumption of fossil fuel like coal, which contains mercury as impurity. Cement production industry has accepted and utilized large amounts of municipal and industrial wastes as secondary raw materials and secondary fuels in Japan. Therefore, Japanese cement industry has played a major role in waste management. Therefore, mercury emission from cement production should be assessed carefully for more reliable mercury emission inventory, especially in Japan. In this paper, mercury emission from cement production based on a mass flow model of cement clinker manufacturing facility would be reported and its uncertainty would be discussed. According to simulation results of the mass flow model, impact of wastes utilization on mercury emission from cement production from 1990 to 2005 would be evaluated.

2. Mass flow model of cement clinker manufacturing facility
2.1 Cement clinker manufacturing process
In this paper, materials supplied to a cement clinker manufacturing are categorized as “natural raw materials (NRM)”, “natural fuel (NF)”, “secondary raw materials (SRM)” and “secondary fuel (SF)”. NRM considered in this paper are limestone, silica sand, and clay. NF are coal, cokes, heavy oil, and light oil. All materials listed as SRM are steelmaking slag (as iron source), coal ash, sludge, residues form nonferrous metal smelting, waste combustion ash, soil from construction demolition. SF are wood wastes, plastic wastes, waste tire, and waste oil. The ratio of utilized wastes (SRM and SF) per unit production of cement clinker from 1990 to 2005 is shown in Figure 1. The ratio of waste utilization in cement clinker manufacturing has increased from 251 kg-waste/Mg-clinker in 1990 to 400 kg-waste/Mg-clinker in 2005. This implies that the impact of wastes on mercury supply to cement kiln has became larger in 2005 compared to 1990 level if mercury content in all input materials (NRM, NF,
SRM, and SF) are assumed to be constant. In this mass flow model, output materials consist of clinker, cement kiln dust (CKD) and exhaust gas. In most cases of cement clinker manufacturing plants in Japan, CKD is mixed into clinker as additives. It should be noted that CKD mixing does not affect product quality and satisfies all quality standard. Therefore, in this mass flow model, all elements contained in CKD are assumed to incorporate into clinker finally.

2.2 Mercury and other elements flow in a clinkering process
All input materials (NRM, NF, SRM, and SF) contain certain amount of mercury as impurity. Other elements (lead, chromium, zinc, and copper) are also considered as impurities. Therefore, these elements are fed into cement kiln via input materials, and then distributed to gas, CKD, and clinker. According to CKD mixing into clinker, all elements would be distributed to gas and clinker product finally. In this paper, all of mercury fed into cement kiln are assumed to transfer to gas phase according to its high volatility. In contrast, other elements are assumed to incorporate into clinker product directly or via CKD.

2.3 Model simulation verification based on element content in clinker product
Content of metal elements (Cr, Cu, Pb, and Zn) in clinker product was calculated by the element flow model as described in previous section. The calculation used NRM, NF, SRM, and SF consumption data and clinker production data for year 2002. The calculation results were compared to real elemental contents in cement clinker product in order to verify simulation results. Although this comparison verify the mass flows of only four tested elements in clinker manufacturing process, not including mercury, it would support partially the certainty of mercury emission from cement production, which is calculated by this element flow model. According to this model calculation, it is possible to evaluate the effect of waste co-combustion in cement kiln on mercury emission.

3. Results and discussions
3.1 Elemental contents in clinker product (Calculation vs Measurement)
Calculated content of metal elements (Cr, Cu, Pb, and Zn) in clinker product was compared to measured data. They were shown in Figure 2. There are non-negligible differences of elemental contents, excluding Cr, between the calculation and measured data for standard clinker sample. However, Cr, Cu, and Zn contents of standard clinker sample are out of the yearly variation range for real clinker product. In contrast, calculated contents of all tested elements are within the yearly variation range. Therefore, Figure 2 suggests that the simulation of tested four elements mass flow in cement clinkering facilities has sufficient certainty. It also supports partially the certainty of mercury emission from cement production, which was calculated by the element mass flow model.

3.2 Impact of waste co-combustion in cement kiln on mercury emission
Mercury is fed to cement kiln via four types of input materials (NRM, NF, SRM, and SF). The share of each input material for mercury emission is illustrated in Figure 3. In 2002 case, atmospheric mercury emission from cement production was estimated to be 6.3 Mg. NRM is the largest mercury donor. It contributes to 64 % of total mercury emission. On the other hand, the share of co-combusted wastes (SRM and SF) on mercury emission is 31%. Therefore, it can be concluded that waste co-combustion in element kiln already has large impact on mercury emission from cement production in Japan. Because SF like wood wastes and plastic wastes has a little impact on mercury emission (only 6 % of mercury emission derived from waste co-combustion), SRM supplied 30 % of mercury emitted to the atmosphere. Figure 4 shows mercury emission from cement production in Japan and the share of mercury sources

Figure 2. Comparison of elemental contents of clinker product between model calculation and measurement (white circle: model calculation, black triangle: measured data of standard clinker sample, black bar: yearly variation of real clinker product)