別紙様式５(Attached Form 5)

学位論文要旨 Abstract of Thesis

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Title of Thesis

Microplastics and plastic additives pollution in open dumping sites soils and environmental samples in Asian regions: Implication for their potential source and exposure

Abstract（within 1600 words）

Plastic production has been increased in global scale since over the past 60 years. Polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC) and polystyrene (PS) are major plastic materials, with PP and PE as the polymer most commonly used in single-used packaging and disposable products. However, ubiquitous distribution of plastic debris and microplastic (MPs) has become a serious environmental issue in the world. Especially, more than 80% of marine debris are considered to originate from Southeast Asian countries. In addition, severe pollution of plastic additives is known to have adverse effects and health risk on the wildlife and human. In recent years, monitoring surveys on plastic debris and MPs residues have been conducted in the environment. These studies reported the present status of MP pollution in air, water, soil, sediment and biological samples collected from European and developed countries, whereas little information is available in Asian developing countries. Further, few studies have been focused on source identification of MPs in the terrestrial environment.

In my *Ph.D* thesis, I focused on the occurrence and distribution of MPs and plastic additives in the environmental samples, such as sediment, road dusts, and dumping site soils (*n*=132), and single-used plastic products (*n*=413) collected from Asian developing countries. The objectives of this study are as follows:

1) To understand the status of MPs and plastic additives pollution in the environment

2) To evaluate a contribution of MPs to the total burden of plastic additives in dumping site soils for the source identification.
3) To analyze specific properties of polymer and organic additives in single-used plastic (SUP) products collected from Asian developing countries.

Information on the comprehensive knowledge of MPs and plastic additives pollution and their source identification obtained in this study play an important role to control their discharge into the environment in Asian developing countries.

As for 'Objective 1', 68 sediments from Myanmar and 10 road dust samples from Thailand collected during 2014 and 2019 were analyzed to understand MPs and plastic additive pollution in Asian developing countries. In Myanmar, high MPs abundance in sediments was found in urban cities, such as Yangon (13,855 pieces/kg), Pathein (12,583 pieces/kg), and Mandalay (11,946 pieces/kg). In contrast, low abundance of MPs was detected in rural towns. Similar results were obtained from MP abundance in road dust from Thailand. The highest MPs abundance was detected in downtown area of Bangkok (7,273 pieces/kg). These results suggest that large population densities, high traffic and high municipal solid waste generation rate may become major MPs sources in the environment. Polyethylene, polypropylene and polyethylene terephthalate with fragments, lines/fibers and films/sheets were major polymers and shape in sediments and road dusts analyzed. This indicates that single-used packaging plastic products derived secondary MPs are potential sources of MPs in Asian developing countries.

Regarding to 'Objective-2', 54 open-dumping site soils collected from Cambodia, India, Indonesia, Laos, the Philippines, and Vietnam were analyzed for MPs. Soil samples were also divided into light (floating) and heavy (sedimentation) fractions by density separation and analyzed for plastic additives. The highest abundance of MP was found in a soil from Cambodia at 218,182 pieces/kg. The median of MP in soils ranged from 1,411 pieces/kg in India to 24,000 pieces/kg in the Philippines, suggesting that dumping sites are a major source of MP into the environment. Polyethylene, polypropylene, and polyethylene terephthalate were dominant polymers in soil samples analyzed. This indicates that daily-used plastic products are main sources of MP in dumping site soils in Asian developing countries.

In plastic additives analysis, five plasticizers DBP, DiBP, DEP, DMP and DEHP, and

an antioxidant BHT were dominant in both of floating and sedimentation fractions. Especially, Bis-(2-ethylhexyl) phthalate (DEHP) was detected as a major additive in soils at mean concentrations of 15,000 ng/g (dry wt.) in floating and 5,700 ng/g in sedimentation fractions. The high concentrations and burdens of phthalates and an antioxidant were detected in floating fraction accounting for 40 to 60% of the total additives in soils. Previous studies on soil pollution have assumed that the organic hydrophobic chemicals analyzed are adsorbed on the surface of soil particles. However, this result indicates that approximately half of the additives in dumping site soils were derived from MP, not soil particle. This strongly suggest that microplastics are potential sources of plastic additive pollution in dumping site soils.

Finally focusing on 'Objective-3', 413 single-used plastic products including plastic bag, food and non-food package, drinking bottle, container, and label film samples collected from Indonesia, Japan, Myanmar and Thailand were analyzed to understand their specific profiles of polymer type and additive concentrations. As the result, three polymers of PE, PP, and PET were mainly identified in plastic products. PE was prominently detected as internal side coupling with other external polymer, whereas PP and PET products were more frequently detected in both internal and external layers. The different polymers used between inner and outer layers of SUP products implies their difficulty in recycling process at high purity.

As for additives analysis, the detection frequency of DEHP was 100% in all target analyzed samples. The highest concentration of DEHP was detected in Myanmar SUPPs PE bags at median 27,000 ng/g which was apparently higher than Japan and Thailand samples. In addition, DBP, DiBP, and DMP levels in Myanmar's products were also greater than those in other Asian countries. The European Union (EU) Committee have decided to restrict the use of six phthalate esters in plastic products in 2020 under the REACH (Restriction, Evaluation, Authorization and Restriction of Chemicals) regulation. The results obtained in this study suggest that several hazardous of plastic additives may be leached or eluted into the aquatic environment in developing countries.

This study clearly suggests that high abundance and ubiquitous distribution of MPs and plastic additives in sediments, road dusts and dumping site soils in Asian developing countries. A single-used plastic products are typical source of MPs and hazardous plastic

additives, and they are leached into the aquatic environment. Also, it was found that insufficient plastic waste management has resulted in severe MPs and plastic additives pollution. Considering these observations, this study provides valuable information how to investigate, control and manage the plastic pollution and their exposure effects on ecosystem in not only Asia but also other regions of developing countries.