主論文の要旨

Redistribution of potentially toxic elements in the hydrosphere after the relocation of a group of tanneries

（皮なめし工場群の移転前後における健康に有害な元素の推移）

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【Background】

The relocation of multiple pollutant sources within a short time frame is a rare event in the world. Such a relocation has been implemented in Hazaribagh, a tannery built-up area with severe pollution, in Bangladesh. These relocations provide a valuable opportunity for practically assessing the associated changes in environmental conditions. In this study, environmental monitoring for a period of 6 years was performed to analyze the distribution of potentially toxic elements derived from tanning process as well as pre- and post-tanning process in the hydrosphere before and after the relocation of tannery operations. According to the actual pollution after the relocation, a potential solution was then considered. The sequence of our results provides novel information for understanding the double-edged effects associated with the relocation of a pollutant source.

【Methods】

Our fieldwork study within a 7-km radius centered on a tannery built-up area was conducted in Hazaribagh. All of the water samples from canals (area b), the confluence (area c) between canals and Buriganga River, and upstream (area a) and downstream (area d) of the confluence before and after the relocation of tanneries were collected in the stationary areas confirmed by using a global positioning system (GPS) (Fig.1A). Since there is no treatment plant, the wastewater from tanneries was directly discharged into the canal before entering Buriganga river. To reduce the heavy environmental pollution, the High Court of Bangladesh decided that the end of 2017 would be the legal deadline for the relocation of tannery operations from Hazaribagh. Water samples (n=203) collected in February 2014 (n=42), February 2015 (n=32) and February 2017 (n=13) were classified as samples before the relocation of tanneries. Water samples collected in January 2018 (n=98) and February 2020 (n=18) were classified as samples after the relocation of tanneries.

Levels of total chromium (t-Cr), lead (Pb), iron (Fe), manganese (Mn), arsenic (As), and barium (Ba) were analyzed using inductively coupled plasma mass spectrometry (ICP-MS). The speciation of trivalent chromium [Cr(III)] and hexavalent chromium [Cr(VI)] were measured by ICP-MS-combined with high-pressure liquid chromatography (HPLC).

The hydrotalcite-like compounds consisting of magnesium and iron (MF-HT) were synthesized and its adsorption capacity were investigated by non-linear adsorption model of Langmuir and Freundlich isotherm model. The application of MF-HT to remove tannery-related toxic elements in selected Hazaribagh canal water (n=10) was performed by 1% (w/v) MF-HT for 1 hour.

【Results】

Our extensive environmental monitoring demonstrated that t-Cr, Cr(III), Cr(VI), Pb, Fe, and Mn, but not As and Ba, were identified as tannery-related elements after the relocation by their

distributions (Fig.1C-I). After identifying the pollutants (Cr, Pb, Fe and Mn) in the hydrosphere, temporal changes of the pollutant levels in the canals (area b in Fig.2A) before and after the relocation were further investigated (Fig.2). The median levels of t-Cr and Cr(III) after the relocation of tanneries were approximately 97% lower than those before the relocation (Fig.2B, C). However, the median level of Cr(VI) after the relocation was up to 30-fold higher than the median levels before the relocation. The median levels of Pb, Fe and Mn (Fig.2E-G) just after the relocation (in 2018) were 57%, 73% and 39% lower, respectively, compared to those before the relocation. However, the median levels of Pb and Fe 2 years after the relocation (in 2020) were 4.5-fold and 3.5-fold higher, respectively, than those just after the relocation. On the other hand, the median levels of both As and Ba were comparably lower than the guideline values of US-EPA throughout our analyses for 6 years.

Since adsorption capacities of Cr(III) and Cr(VI) and Fe by MF-HT have been reported in our previous studies, the capacity of MF-HT to adsorb other tannery-related elements (Pb and Mn) was newly investigated in this study (Fig.3). After confirming the adsorption abilities of MF-HT, it was investigated whether MF-HT could actually remediate the elemental pollution in the canal water samples (Fig.4). In all of the treated canal water samples (n=10), the concentrations of Cr, Pb, Fe and Mn were decreased to below the guideline values of US-EPA.

【Discussion】

Since there were approximately 97% decreased levels of t-Cr and Cr(III) in canal water samples after the relocation compared to those before the relocation, the relocation of tanneries was partially successful from the viewpoint of Cr(III) water pollution. However, the median Cr(III) level after the relocation of tanneries remained higher than the US-EPA guideline value. These results suggest not only continuous water pollution of Cr(III) after the relocation but also no further improvement of the Cr(III) pollution for 2 years after the relocation.

The situation of Cr(VI) pollution after the relocation was more serious than that of Cr(III). The pollution of Cr(VI) in canal water after the relocation progressed over time. These results suggest the conversion of Cr(III) to Cr(VI) in nature. It has been theoretically shown that Cr(III) in soil polluted by tannery wastewater represented as tannery sludge could be oxidized through interaction with MnO_2 in an aerobic condition. Since the converted Cr(VI) is highly soluble, it could migrate into the aqueous environments from the soil. It has been experimentally shown that Cr(III) in tannery sludge could be converted to Cr(VI) under various conditions pertaining to the natural environment. The oxidization of Cr(III) in the bed and bank soils of canals via increased aeration of the soils by the decreased canal water level through the decreased number of tanneries associated with the relocation may actually increase the level of Cr(VI) in the hydrosphere of Hazaribagh.

Pb, Fe and Mn were then focused on as representative pollutants caused by the pre- and post-tanning processes. Median levels of these elements were temporarily decreased just after

the relocation. However, the median levels of Pb and Fe 2 years after the relocation were about 3.5 to 4.5-fold higher than those just after the relocation. Considering the report of an increase in tanneries handling the processes other than tanning after the relocation (in 2019), these results suggested that recent elemental pollution caused by the pre- and post-tanning processes was exacerbated.

Finally, remediation of polluted canal water was attempted by using MF-HT. The MF-HT could also remediate all of the tannery-related toxic elements in the raw canal water samples to levels lower than the US-EPA guideline values. The daily expense of raw materials for remediating most of the elements from the wastewater could be estimated as less than 2 cents per day. Since a plan for the establishment of residential land in Hazaribagh is progressing, the MF-HT could be a powerful tool for decreasing the health risk through environmental remediation in a vacant lot after the relocation of tanneries.

【Conclusion】

The drastically decreased level of Cr(III) after tannery relocation indicates a beneficial effect of tannery relocation. In contrast, the strikingly increased level of Cr(VI) indicates a harmful effect of the relocation. Our fieldwork study newly suggests promotion of the conversion of anthropogenic Cr(III) to Cr(VI) in nature. Considering the toxicities caused by Pb, Fe and Mn in addition to Cr, health hazards in tannery built-up areas remain even after the relocation of tanneries. This study suggested that the use of MF-HT could be effective for solving the problems of environmental pollution and health hazards remaining after the relocation of pollutant sources.