

論文の内容の要旨

Mercury exposure among general Pakistani population — Personal care products as a source of excessive exposure.

(パキスタンの一般公衆の水銀曝露—過剰曝露源としてのパーソナルケア製品)

サディア カンワル

Mercury is one of the persistent and toxic elements to which humans are exposed from both natural and anthropogenic sources. The adverse health effects of mercury on humans depend on its chemical forms, concentrations and vulnerability between exposed subjects. Due to its long range transport, biogeochemical cycling and toxicity, a mercury programme was established by the governing council of United Nations Environmental program (UNEP) to review the strategies that had been taken in the past for reducing mercury levels from the environment. Governing council decided after extensive review that old strategies was not sufficient to reduce mercury levels and decided to take actions internationally. For this purpose, intergovernmental negotiating committee (INC) was established in 2010 and concluded its sessions in 2013 by agreement on the Minamata Convention on Mercury. The main objectives of the convention were; i) to protect the human health from the release of mercury and its compounds particularly generates due to anthropogenic activities, and ii) to set out the range of measures for the mercury levels reduction from the environment. Pakistan is one of the signatories of Minamata Convention.

Despite its serious environmental concerns, studies on mercury exposure levels in Pakistan are quite limited. Based on this fact, the present study was conducted to assess the current mercury exposure status of Pakistani urban dwellers to check contamination levels by using hair samples. Current mercury exposure levels of Pakistani individuals will be valuable as a background information in the future for the evaluation of effectiveness of the Convention. The second major objective of this study was the characterization of high mercury concentrations in excessively exposed subjects with speciation analysis and mercury stable isotopes analysis. The assessment of health risks of Pakistani individuals due to the use of mercury containing products was also one of the objectives of this study.

The ethical review committee of The University of Tokyo approved this study. To achieve the objectives of the study, hair sampling was carried out in six major cities (Lahore, Islamabad, Karachi, Peshawar, Sialkot and Rawalpindi) of Pakistan. Hair samples were collected from the 291 subjects (136 males and 155 females), properly labelled, stored in a polythene bags and shipped to Japan for further analysis. Questionnaires data were also filled by the subjects during sampling in which the information of subjects related to age, place of residence, occupation, education, smoking habits, use of personal care products (PCPs), use of skin lightening creams, soaps and its frequency, fish consumption, types of fish consumption were also collected. The informed consent of participating in this study was also obtained from the subjects.

Total mercury concentrations were measured in the hair samples of 291 subjects. Statistical analysis was performed between hair mercury concentrations of subjects and other demographic and life style factors to identify the source of variations of mercury exposure. Hair total mercury concentration data of the subjects were categorized into two groups on the basis of percentile distribution. Subjects with hair mercury concentrations below 95th percentile were named as non-excessive exposure group (NEEG), and subjects with hair mercury concentration above 95th percentile were named as excessive exposure group (EEG). According to percentile distribution data, EEG included the subjects had hair mercury concentrations above 11 µg/g. In addition, NEEG included the subjects had hair mercury concentrations below 11 µg/g. Overall, results showed that 70 % of the subjects of this study had hair total mercury concentrations below 1 µg/g, the permissible limit of mercury in hair samples recommended by United States Environmental Protection Agency (USEPA). In addition, EEG included 14 subjects had hair mercury levels ranged from 11.1 µg/g to 565 µg/g that can pose significant health risks to the exposed subjects. NEEG included 277 subjects had hair mercury levels ranged from <0.016 µg/g to 10.2 µg/g. Association of EEG/NEEG with categorical variables including gender, education, occupation, place of residence, smoking, passive smoking, use of PCPs, use of skin lightening creams and soaps, usage frequency of skin lightening creams and soaps, fish consumption and its types were also tested by statistical analysis. Use of skin lightening creams, soaps and its usage frequency were found as statistically significant factor associated with the categorization of EEG and NEEG subjects. Therefore, it was hypothesized, that EEG subjects were exposed to excessive mercury levels via the use of skin lightening creams and soaps.

Speciation analysis found that hair samples of EEG subjects had excessive concentrations of inorganic mercury (4.16 to 113 µg/g). Methylmercury concentrations were considerably low (<0.005-0.219 µg/g) and ethylmercury species were not detected (<0.005 µg/g) in the hair samples of EEG subjects. In addition, some unidentified mercury species (0.01-0.73 µg/g) were also detected in the hair samples of EEG and its retention time was not matched with the retention time of methyl, ethyl and phenyl mercury. Results of speciation analysis support the hypothesis that EEG subjects were exposed to excessive levels of mercury probably by the use of skin lightening creams and soaps. Because inorganic mercury salts are commonly used in the creams and soaps in some of the African and Asian countries.

In this study, skin lightening cream and soap samples reported by EEG subjects in the questionnaires were also collected from Pakistan. These samples were measured for the total mercury concentrations to analyze either these products contained mercury or not. It was found from the results that 3 skin lightening cream samples out of 10 analyzed products had total mercury concentrations ranged from 0.0040 to 3.4 %, which was considerably high compared with the maximum allowable limit of 1 $\mu\text{g/g}$, according to United states Food and Drug administration (USFDA). Mercury stable isotope ratios were measured in the hair samples of EEG subjects and skin lightening cream samples used by the subjects. Results showed that mercury stable isotope ratios of hair of one of the EEG subject was matched with the skin lightening creams isotope ratios reported by the subject. Moreover, hair isotope ratios of the other 3 EEG subjects did not match with skin lightening creams isotope ratios reported by those subjects. The results of mercury stable isotopes analysis indicated that the skin lightening cream samples reported by the EEG subjects are not the sole source of excessive inorganic mercury exposure among EEG subjects except in one subject. It is highly probable that EEG subjects (in which hair mercury stable isotope ratios did not match with skin lightening creams isotope ratios) were exposed to other inorganic mercury exposure sources like the use of other creams and soaps or any other environmental (atmospheric particles, dental amalgams, occupational activities etc) sources.

Skin lightening cream samples (detected with mercury concentrations up to 3.4 %) were also checked for the possible health risks to which Pakistani people can be exposed after using these products. For this purpose, HQ was estimated for the assessment of health risks which showed that 2 skin lightening cream samples had HQ greater than 1, indicated that the use of these skin lightening creams may pose significant health risk to its users.

It is recommended in future studies to explore other possible sources of inorganic mercury through which people in Pakistan are exposed to excessive levels. Identification of inorganic mercury exposure sources is essential to reduce excessive levels of mercury from the environment. In addition, identification of unknown mercury species in the hair samples of EEG subjects is also recommended for future studies. Moreover, other skin products, particularly creams and soaps, sold in Pakistan must be analyzed for mercury concentrations to assess the potential of health risks. Data is limited to total mercury concentrations in the biological and environmental matrices in Pakistan. So, future studies related to the speciation analysis in the biological samples (i.e. blood, urine) of Pakistan must be carried out that will be helpful in health risk assessment of individuals in case of excessive mercury exposure.