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Pharmaceuticals and personal care products (PPCPs) and anthropogenic gadolinium in groundwater in central Tokyo

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Together with an increasing concern about water scarcity caused by climate change, groundwater can serve as a supplementary source of water in Tokyo for various purposes including drinking, landscaping, water in emergency, water for spraying to mitigate heat island in summer. However, studies have shown that some of groundwater in Tokyo was polluted by various contaminants including nitrogen, bacteria, organic carbon, and perfluorinated compounds, some of which originate from domestic wastewater. Therefore, for the protection of groundwater quality, assessing groundwater pollution by domestic wastewater in the city-wide scale is needed.

With that aim, we measured pharmaceuticals and personal care products (PPCPs) and anthropogenic gadolinium, which are considered to be specific to domestic wastewater. Measured PPCPs include carbamazepine, a quantitative tracer of sewage in groundwater reported in overseas, and crotamiton, which has been reported to be conservative and hardly attenuated in soils in Japan. Anthropogenic gadolinium, which supposedly originates from contrast media for magnetic resonance imaging (MRI) in hospitals, has also been reported as a conservative tracer of wastewater in groundwater.

Totally 50 groundwater samples were taken between October to December 2007 in 19 wards in central Tokyo: 32 sites from unconfined aquifers with approximately 10-30m of depth; 16 sites from confined aquifers with approximately 30-500m of depth; 2 sites from springs on the Musashino Terrace. For PPCPs measurement, samples were filtered (0.7um), spiked with deuterated surrogates, concentrated with solid phase extraction, purified with 5% water-deactivated silica gel column chromatography, and analyzed by GC-MS. For measurement of anthropogenic gadolinium, samples were filtered (0.45um), added with 60% HNO3 (1%(v/v)), spiked with bismuth as an internal standard. Gadolinium was measured together with the other rare earth elements by ICP-MS. Concentration of anthropogenic gadolinium was calculated as difference of total gadolinium measured by ICP-MS and geogenic gadolinium estimated with samarium and terbium when ratio of total- to geogenic gadolinium exceeds 1.3.

PPCPs were detected in 21/32 sites (66%) from unconfined aquifers, 2/2 sites (100%) from springs, and 7/16 sites (44%) from confined aquifers. More detection in unconfined aquifers and springs than confined aquifers indicates that groundwater was polluted by domestic wastewater near the ground surface. Detection in confined aquifers was mainly located near the border between the Musashino Terrace and the Tokyo Lowland. We estimated that the Tokyo layer, which is the main confined aquifer in that area, was polluted by the infiltration of upper unconfined groundwater. Concentration of PPCPs in groundwater was 1-2 orders lower than that of sewage influent, while in some groundwater PPCP concentration was comparable with sewage influent. Among six PPCPs measured, carbamazepine and crotamiton were most frequently detected (19/50 sites and 18/50 sites, respectively). PPCPs were detected in four sites where E. coli was detected. It was considered that PPCPs were more widely detected than E. coli because E. coli is more easily attenuated by sorption to soil surface and die-off in groundwater.

Anthropogenic gadolinium was detected in seven unconfined aquifers and one confined aquifers. The distribution of PPCPs and anthropogenic gadolinium was quite different: at only three sites both compounds were detected. E. coli was detected in none of anthropogenic gadolinium-positive sites. Those differences may arise from the distribution of the compounds in wastewater. PPCPs are considered to be widely contained in domestic wastewater, while it was reported that anthropogenic gadolinium was highly abundant in effluent from MRI-equipped hospitals. Therefore, it was considered that anthropogenic gadolinium-positive groundwater may have a strong influence by effluent from MRI-equipped hospitals.

Keywords: groundwater pollution, domestic wastewater, pharmaceuticals and personal care products (PPCPs), gadolinium, E. coli