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Title: A field survey on elution of lead and nickel from taps used in homes and analysis of product test results

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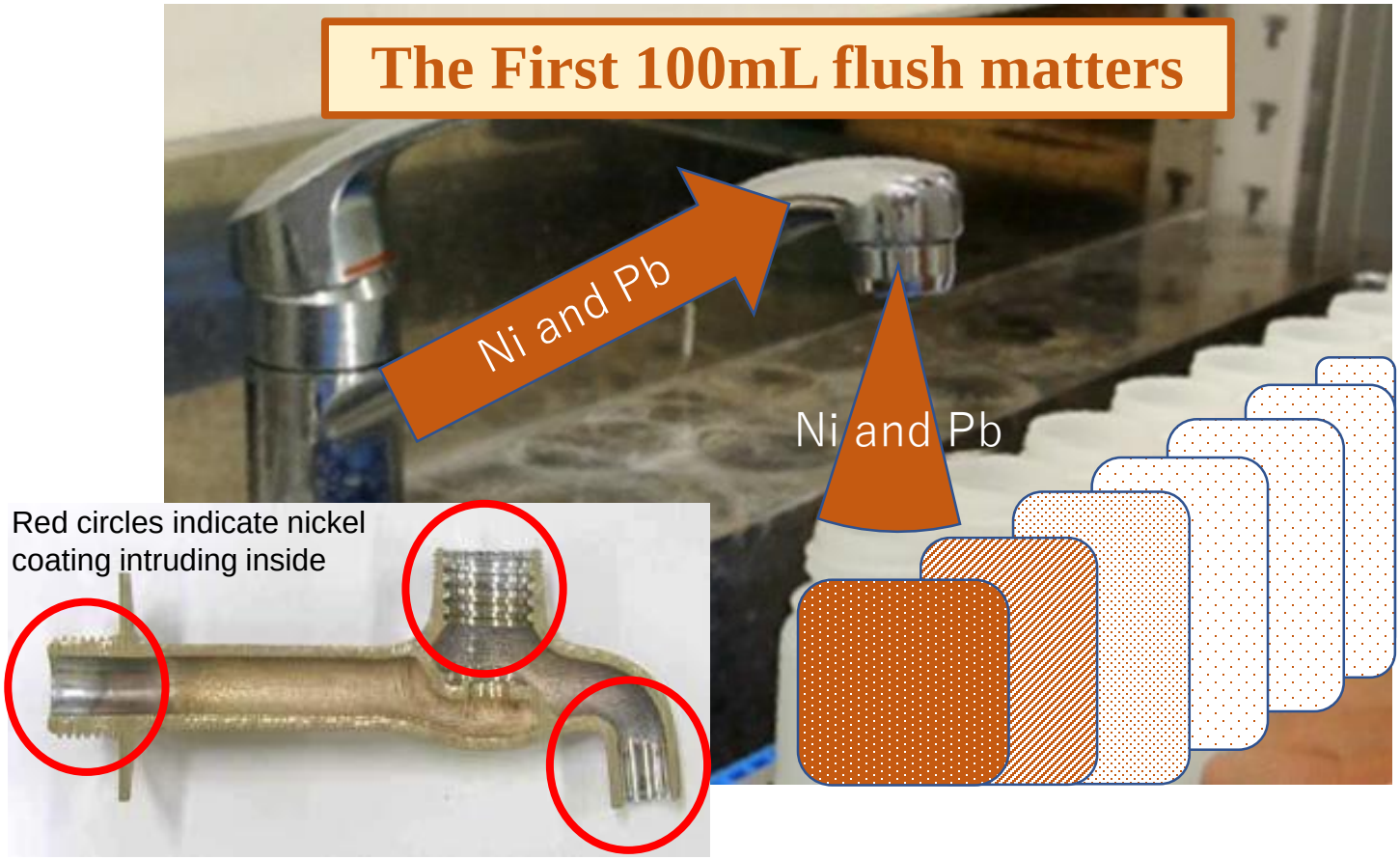
ABSTRACT

The elution of lead, and nickel from water supply devices into water is a potential health concern. This study was performed to examine the actual concentrations of nickel and lead in the water from taps in homes and offices, focusing on the differences between first flush and fully flushed water. The water quality management target value and water quality standard in Japan specify nickel and lead concentrations in drinking water < 20 and < 10 µg/L, respectively. Nickel concentration in the first flush water (100 mL) from 110 household taps revealed 22 cases (20%) > 20 µg/L, while the fully flushed water satisfied the standard after running 5000 mL of water. The nickel concentration decreased gradually in sequential sampling of each 100 mL from the taps. Lead concentration in the first flush water exceeded the standard in 32 cases (29%), while the fully flushed water was below the target value. The concentration in the first flush water tended to decrease with time since the tap installation, and this was significant after 10 years for nickel but not significant for lead. It is important to flush retained water out of the tap after several hours without use. No significant correlation was found with the volume of the test faucet in the market, but bronze-based products showed higher nickel concentrations than brass and plastic products.

Keywords: first flush, elution, home water supply device, inorganic chemical



・20% and 29% of the first flush (100mL) in 110 household water taps exceeded 20 μg/L of Ni and the standard of Pb, respectively.

Graphical abstract

Highlights (for review : 3 to 5 bullet points (maximum 85 characters including spaces per bullet point)

Highlights

- Ni concentration in first 100-mL flushes from 20% of the household taps exceeded 20 µg/L.
- Pb concentration in first 100-mL flushes of 29% of the taps exceeded the standard.
- Ni and Pb concentrations did not necessarily decrease over time.
- Bronze-based faucets showed greater Ni elution.

1. INTRODUCTION

The elution of lead, chromium, and nickel from water supply devices into water is a potential health concern. Elution of nickel has long been a concern from the perspective of not only direct dermal contact with faucets but also indirect contact through tap water (Andersen *et al.* 1983; Gammelgaard *et al.*, 1985). The World Health Organization (WHO) Guidelines for Drinking Water Quality, 4th edition (WHO, 2011), states that nickel present in tap water is eluted from new nickel- and chrome-plated faucets or pipes and joints made of stainless steel. The evaluation document of nickel published by the Food Safety Committee of Japan in July 2012 stated that the main adverse effect of nickel on the human body is allergic contact dermatitis (FSC, 2012). With regard to genotoxicity, in vitro experiments have shown that nickel induces DNA damage, gene mutation, and chromosome aberrations in cultured mammalian cells. However, there have been no reports regarding in vivo experiments to examine the effects of nickel on gene mutation, so the carcinogenic potential of nickel by oral exposure remains unknown. Therefore, it is appropriate to calculate the tolerable daily intake (TDI) of nickel in relation to non-carcinogenic toxicity. According to the nickel evaluation document mentioned above, the TDI of nickel in non-carcinogenic drug toxicity tests was evaluated as follows. Administration of nickel via drinking water was performed to test nickel dermatitis on an empty stomach. It was then checked whether the maculopapular rash and dermal damage became enlarged or worsened. The lowest observed adverse effect level

(LOAEL) was determined as 12 µg/kg body weight/day. The TDI of 4 µg/kg body weight/day was derived from the LOAEL divided by an uncertainty factor of 3 because of the uncertainty of the experimental results. The water quality management target value for nickel in Japan is 20 µg/L (Ministry of Health, Labour and Welfare [MHLW], 2016), which conforms to the EU directive value (EC, 1998) but is lower than the WHO guideline value of 70 µg/L (WHO, 2011). However, the water quality and elution of nickel from water supply devices are currently not determined as national enforceable standards in Japan because nickel is rarely found in raw water and is only sometimes found in tap water. One of 2540 tap water samples exceeded the target value in 2015, while none exceeded the target in 2016, 2017, or 2018 (JWWA, 2018). However, nickel may originate from water supply devices, but the samples tested for water quality examination were fully flushed waters, and the quality of standing water that may be consumed has not been fully investigated. In a nationwide survey conducted in Germany (Volker *et al.*, 2010), 10.9% of all samples exceeded 2 µg/L, but it was not reported whether the excess was related to the water retained in the water supply devices. Sorlini *et al.* (2014) reported that 11% of the water samples collected at taps contained valves with nickel levels > 20 µg/L suggesting the release of nickel from the valve material.

The health effects of lead have been a matter serious concern among international and national organizations for many years. The WHO (2011), International Association for Research on Cancer (IARC, 2006), Centers for Disease Control (CDC, 2019), Environmental

Protection Agency (EPA, 2016), Food and Drug Administration (FDA, 1993), and National Toxicology Program (NTP, 2012) conducted risk assessment and established standards for lead. While the main source of lead exposure is house dust due to airborne particles in Japan (Yoshinaga, 2012), Renner (2010) pointed out that drinking water is an overlooked source of lead as the water quality fluctuates according to both sampling methods used and sampling season. A nationwide survey conducted in Germany (Volker *et al.*, 2010) showed that tap water exceeded the EU directive for lead level by 4.7% (10 µg/L). Lim *et al.* (2013) reported that first flush samples had significantly higher lead levels than fully flushed samples and lead concentration exceeded 10 µg/L in some areas, but not in others. Sorlini *et al.* (2014) suggested that lead was released from internal water piping networks in buildings. Harvey *et al.* (2016) reported that the major source of lead contamination was plumbing components followed by kitchen faucet components. Etchevers *et al.* (2015) performed a nationwide cross-sectional survey in 2008 – 2009 in France, and confirmed that household dust and tap water made the largest contributions to blood lead levels. Lead concentrations in tap water > 5 µg/L were positively correlated with the blood lead levels in children drinking tap water. Deshommes *et al.* (2013) discussed how tap water influences children's blood lead levels based on monitoring data and uptake model simulations. Since the deterioration of water quality due to water source changes occurred in Flint, Michigan, in the USA, there have been a number of studies of the water supply system and lead exposure especially in children in the

area (Deshommes *et al.*, 2016; Kennedy *et al.*, 2016; Lytle *et al.*, 2019). For example, Kennedy *et al.* (2016) reported that blood lead levels were significantly increased due to lead exposure in supplied water during the period of maloperation of Flint's water supply in 2013 – 2016. The characteristics of particulate matter were also examined in domestic water (Clark *et al.*, 2014), as well as water passing through pipelines in buildings (Chang *et al.*, 2019; Kinoshita *et al.*, 2016) and on campuses (Chang *et al.*, 2019). Rosen *et al.* (2017) and Bradley *et al.* (2018) showed that metal contamination from pipes and organic substances, including bacterial growth in the distribution network, are correlated with water quality at the faucet. Dore *et al.* (2019) investigated water quality and flushing to minimize lead release after partial lead service line replacement, and reported that high-velocity flushing before stagnation was effective. The US EPA has set the maximum contamination level of lead at 50 µg/L, which is referred to as the treatment technology-based action level (EPA, 2016).

MHLW determined the water quality standard as < 10 µg/L in 1994 and a moratorium for adoption was settled because a period of 10 years will be needed to replace lead pipes and faucets. In 2019, MHLW noted that 2.59 million houses are still connected to lead pipes and an urgent countermeasure for replacement was requested. Around this time, FSC of Japan attempted to revise the toxicological evaluation of lead mainly in bottled drinking water. We performed a field study of the elution of nickel and lead from faucets in ordinary households and office buildings in Japan. The objective was to better understand actual exposure to

77 inorganic materials from taps, because people tend to drink or use tap water after a period of
78 disuse, thus allowing water to sit in the pipes. In addition, to understand the characteristics of
79 nickel elution, the results of the elution tests were analyzed, and the materials and types of
80 faucets as well as elution of nickel were examined.

81 ## 2. MATERIAL AND METHODS

82 ### 2.1 Field study

83 *First flush and fully flushed water sampling*

84 The field study involved collecting tap water samples under two conditions for each faucet,
85 requesting ordinary households and offices to collect water, in September 2017. Here, we use
86 the term "tap" in reference to the faucet connected to the water supply network, while the
87 device itself is referred to as a "faucet." Samples were collected from a total of 110 taps in
88 Japan, including Kanto (east), Kansai (west), Tohoku (northeast), Hokkaido (north), and
89 Kyushu (south) areas. Water was collected at ordinary households and offices under two
90 conditions into 100-mL polyethylene bottles as follows: 1) first flush, first 100 mL of water
91 drawn from a tap that had not been used overnight (> 6 hours); and 2) fully flushed water, 100
92 mL of water sampled from the same tap as in 1) after passage of 5000 mL of water. A
93 questionnaire regarding the tap was enclosed to acquire data, including time to obtain the
94 water, location of the faucet, type of faucet, number of years after installation of the faucet,

and manufacturer of the tap. Photographs of the types of some taps are shown in Fig. S1. Samples were analyzed once and quality control was performed every 10 samples.

Sequential sampling

Successive samples of 100 mL of tap water were collected after one night. A total of 20 bottles each holding 100 mL were taken. A photograph of the sampling procedure is shown in Fig. S2.

Time interval sampling

For selected taps in which nickel was detected, first flush of 100 mL was sampled after 1, 2, 4, 6, and 8 hours without use of the tap. After water sampling, 5000 mL of water was flushed to prepare for the next water sampling. This test was conducted twice in two faucets in which the first flush showed higher concentrations.

Chemical analysis

Collected samples were examined for nickel (Ni), lead (Pb) and chromium (Cr) by inductively coupled plasma mass spectrometry (ICP-MS) (7500cs; Agilent Technologies, Santa Clara, CA, USA). The standard solutions were as follows: nickel standard solution (Ni 1000, containing 1000 mg/L of nickel; Wako, Osaka, Japan), chromium standard solution (Cr 1000, containing 1000 mg/L; Wako), lead standard solution (Pb 1000, containing 1000 mg/L; Wako), tuning solution (Ce, Co, Li, Mg, Tl (thallium), Y (yttrium) 1 μg/L, HNO_3: 2 wt%, 500

mL), nitric acid (for fine analysis: Wako), pure argon G3 (purity ≥ 99.999%; Taiyo Nippon Sanso Co., Tokyo, Japan), and pure helium G1 (purity ≥ 99.99995%; Taiyo Nippon Sanso Co.). Vials used for measurement were dipped in nitric acid solution for 24 hours or more and then washed with ultrapure water. For sample pretreatment, 1 mL of HNO_3 was added to every 100 mL of sample and then stored in a refrigerator for 1 day or longer. A calibration curve was produced for each measurement. For measurement, the quantification range for low concentration was used. Samples exceeding the quantification range were quantified using the calibration curve of higher concentrations. Samples were analyzed once and quality control was performed every 10 samples.

2.2 Examination of faucets by trial purchase and analysis

The National Institute of Public Health (NIPH) is responsible for market watch of water supply devices by trial purchasing from the market. Fifty-four water supply devices for daily use, such as faucets, were purchased from commercial sources according to the share in the Japanese market to test the strength, material, and configuration following the waterworks law. Faucets were divided into two groups. NIPH performed elution tests of water supply devices from 2012 to 2017. The purchased devices were subjected to an elution test with or without conditioning (Fig. S3), according to the protocol of the MHLW Notice 111, "Standard Test Related to Water Supply Devices and Materials" [11] with some modifications.

According to the protocol, when the inner volume of a water supply device is < 1 L, the results are adjusted to 1 L to calculate a normalization factor. In this study, however, we did not make this adjustment to properly evaluate the elution properties. The analysis was performed by ICP-MS, and the lower limit of detection for nickel was 0.5 μg/L.

3. RESULTS

3.1 Field study and discussion

First flush and fully flushed water sampling

The nickel and lead concentrations in samples No. 1 to 110 are shown in Fig. 1. The measurement results indicated that the water quality management target value (20 μg/L) in the first flush was exceeded in 22 of 110 cases. On the other hand, after discharging ≥ 5000 mL of water, all samples satisfied the target value. With regard to lead concentration, 32 of the 110 first flush samples from the taps exceeded the standard value. However, all fully flushed samples obtained after flow of 5000 mL of water satisfied the standard. Samples in which nickel showed a high concentration also tended to contain detectable levels of lead. There were two cases in which chromium was detected at 1 μg/L, which was the lower limit of detection, but fully flushed water was below this level.

The relations between nickel and lead concentrations in first flush and the number of years since installation were investigated (Fig. 1). The samples were obtained from 64 taps in use

for ≤ 10 years and 46 taps in use for > 10 years. The water from the newer faucets had significantly higher nickel concentrations ($p < 0.05$), while lead concentration was not significantly higher in water from new taps ($p = 0.14$).

Figure 2 shows the relationship between nickel and lead in first flush water samples on a log-log plot. They were highly correlated ($r = 0.59$, $n = 110$, $p < 0.01$) when values below the limit of quantification were substituted by 0.5 µg/L, i.e., half the value of the quantification limit (1 µg/L).

Sequential sampling

Water samples of 100 mL were collected continuously from 11 selected taps that had shown high nickel concentrations in the first flush and fully flushed water samples and were analyzed up to 600 mL for nickel and lead (Fig. 3). Although the nickel concentrations were at or above the water quality management target value for 0 – 100 mL (first flush) and/or 100 – 200 mL, values after flushing of 300 mL were below the target value. All of these taps showed lead concentrations no higher than the standard value. For some taps (Taps A and E), however, the concentration of lead decreased more gradually compared to that of nickel. It was inferred that the drop in concentration was slow because the lead was derived from the pipes rather than the faucet, although the materials and volume of the taps and pipes were not clear as they are in daily use in houses. Chromium levels were also investigated (Fig. S4), but the concentrations were low compared to the standard value (20 µg/L). The first flush of 100

or 200 mL of water contained higher concentrations of nickel and lead and should not be used.

Interval sampling

The effects of retention time on the concentrations of nickel and lead in the first flush were investigated in two taps (Taps I and K), as shown in Fig. 3. The results for nickel and lead are shown in Fig. 4. In both taps, the nickel concentration in the first flush increased with retention time. For Tap K, the concentration came close to the target value with several hours of retention. Therefore, the concentration was thought to increase even after one night in daily use.

3.2 Trial test and discussion

Faucet manufacturers

The values of nickel determined by analysis with or without conditioning for each manufacturer are shown in Fig. 5. The products by companies C and E tended to elute nickel more than the others. The main materials of the faucet body produced by both companies C and E were brass, bronze, and resin, but among them faucets made of bronze tended to elute nickel.

Inner volume and type of faucet

The relations of the unit void volumes (total water-holding volume of internal components)

were investigated. The average concentrations of eluates for faucets with volume ≤ 100 mL were 990 µg/L without conditioning and 858 µg/L with conditioning, both of which were higher than the average values for faucets with volume > 100 mL, i.e., 592 µg/L without conditioning and 266 µg/L with conditioning. It was suggested that nickel tended to elute from the faucets. The relation between inner volume of faucets and concentration of nickel eluted into the water is shown in Fig. S5.

Main material of the faucet

Faucets in the trial purchase study were divided into three types according to the main material of the main body. As shown in Fig. 6, the average nickel concentration for faucets made of brass was 126 µg/L and the average value after conditioning was 8 µg/L. The average value for faucets made of plastic was 108 µg/L and the average value after conditioning was 7 µg/L. The average value for faucets made of bronze was 1393 µg/L and the average value after conditioning was 79 µg/L. Faucets with bronze as the main body material were manufactured by several companies, and these tended to elute nickel. For nickel-coated faucets, it is difficult to control coating even in products of the same lot (Fig. S6).

4. DISCUSSION

The quantity of nickel eluted into the water from faucets was reported to decrease over time

after installation (WHO, 2011). These faucets may have had higher nickel concentrations when they were first installed. On the other hand, old faucets may have contained higher concentrations of lead, which has remained until the present. It is important to note that faucets ≥ 10 years old did not necessarily show a low nickel or lead concentration in first flush water samples. There seemed to be two main reasons for this. The first is elution, where new materials elute heavy metals and the concentration gradually decreases near the surface, and the second is the change in surface condition, such as deposition or the development of a passive layer covering the surface of the material and further decreasing elution. Orthophosphoric acid is known to cover the surface of faucets and has been used in water treatment in the USA; however, in Japan, water suppliers use soda to raise the pH of water (Hamamoto et al., 2007).

Other factors, such as pH, manufacturer, and type of tap, were also analyzed (Fig. S7, S8, and S9, respectively). However, as shown in Fig. S7, pH ranged from 6.9 to 7.9 and showed no direct correlation with elution profile. As there were large degrees of variation in pH and concentration among taps, these factors did not show significant correlations with nickel and lead concentrations. We examined regional variation, but the variation in data among areas was so large that the differences were not statistically significant, i.e., average of northeast = 31.2 μg/L, east = 32.8 μg/L, and west = 26.8 μg/L, with SD = 81.1, 79.3, and 78.0, respectively.

It was assumed that the water contained higher levels of heavy metals eluted from the faucet or pipe materials in the vicinity. Other conditions, such as retention time, temperature, and water quality, may also have affected the elution of these metals.

Although the variation was quite large, the average eluate concentration was lower for faucets with greater inner volume. Without conditioning, both low and high concentrations were mixed regardless of the contact volume, and concentration values were spread over a wide range. Small single faucets showed higher nickel concentrations in the water than mixed faucets with larger volumes.

Faucets made of bronze showed significantly higher concentrations of nickel in the water than faucets made mainly of brass and plastic; however, the original contents of bronze faucets were not clear. The concentration of nickel for mixed faucets tended to be lower than for standard faucets.

Schock and Neff (1988) reported that brass valves and fittings more readily eluted lead from their components. However, in this study we did not collect samples sequentially along the background pipe system. Lei *et al.* (2018) reported that plumbing materials should be monitored for lead release in new premises. Masters *et al.* (2016) also reported that temperature markedly influences lead concentrations in water, because the solubility depends on the temperature of the water. As these factors were not examined in the present study, they must be monitored thoroughly over a longer duration taking into consideration the location,

size, temperature, and length of the target part of the material in the plumbing system within the building.

There is also concern regarding lead eluted into the water supply distribution pipes consisting of many parts, including valves and fittings, as described in the Supplementary Information. In Japan, the official sampling method for lead is to sample water 1) after a full flush, 2) wait for 15 minutes, and 3) take a sample from a 5000-mL pool.

Our research focused on taps by assuming that distribution pipes in houses in Japan are not a major source of nickel. Sequential sampling in normal houses was performed from first flush to 600 mL in increments of 100 mL, which is larger than the average inner volume of a water supply faucet. However, we felt that this sampling protocol closely represented the real situation. This is a type of citizen science, as reported by Redmon *et al.* (2020), who used water samples collected under conditions of normal use mostly by users. This type of trial may confirm the actual exposure to contents through water supply devices in addition to the outlet of water supply treatment plants.

5. CONCLUSIONS

Nickel and lead were eluted in relatively high concentrations in the first flush. Nickel and lead were considered to be eluted from the taps installed and used in normal houses. The water quality management target value (< 20 μg/L) was exceeded in the first flush for 22 of 110 taps examined in the field study. However, the management target value was mostly satisfied in

fully flushed water after flowing 5000 mL of water. Faucets in which the water quality management target value was exceeded in the first flush showed values below the water quality standard after flowing 300 mL of water. With regard to the effects of retention time, nickel concentration of the first flush could come close to the management target value after several hours of disuse.

The standard value for lead in first flush water was exceeded in 32 faucets. However, all faucets satisfied the water quality standard in fully flushed water. Nickel and lead concentrations in first flush water were correlated in the field survey.

The number of years since installation was believed to be inversely related to elution of metals. However, the results of this study showed that there was a significantly higher nickel concentration within 10 years, but no significant relation was observed between lead concentration and number of years since installation. As nickel showed a greater tendency to elute from faucets made of bronze, the results suggested that the material from which the faucet is made may be the main factor related to metal elution into water.

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SUPPLEMENTARY MATERIALS

Your proofreading of references is not required.

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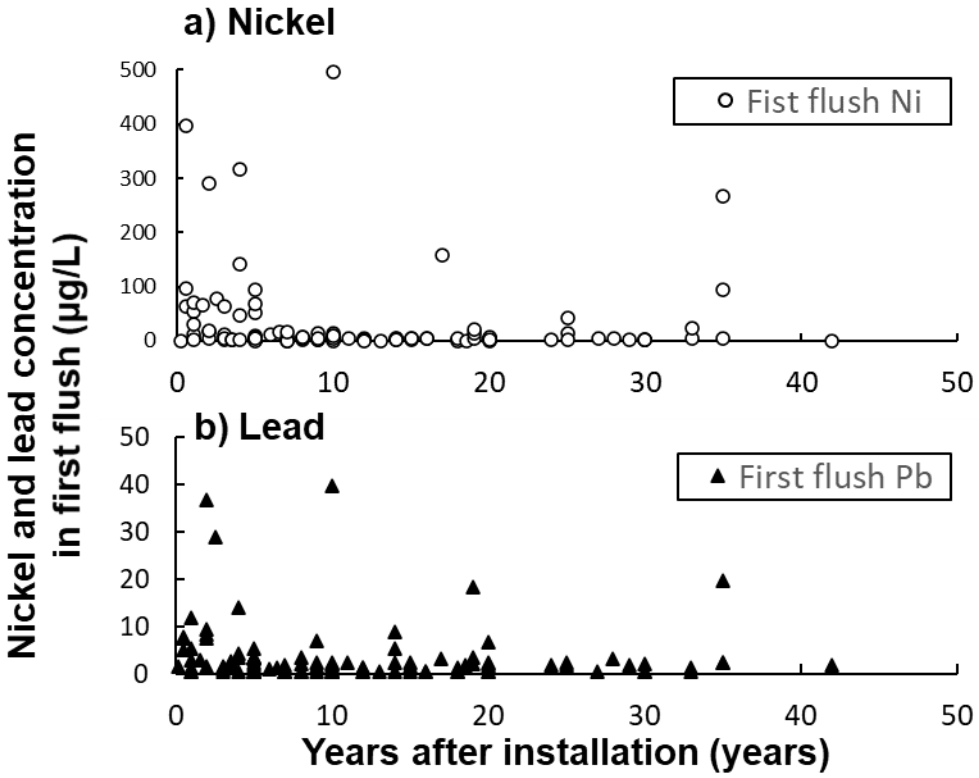

Fig. 1 Nickel (a) and lead (b) elution concentrations of the field survey taps and number of years since installation

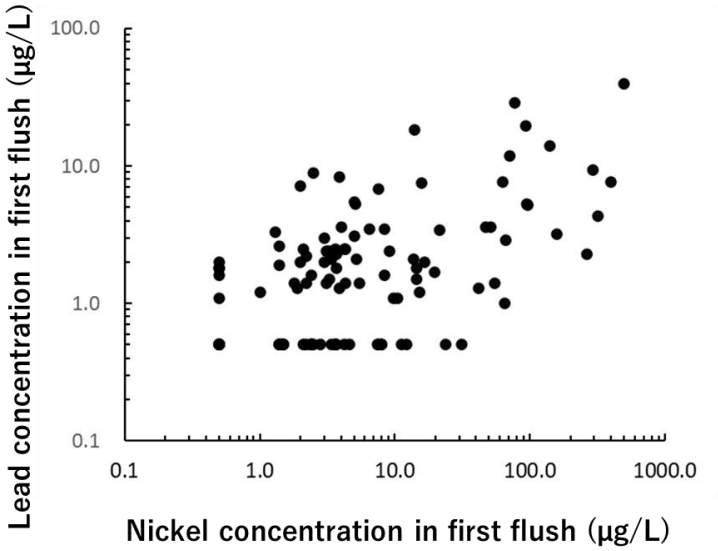

Fig. 2 Relationship between nickel and lead in first flush in field taps

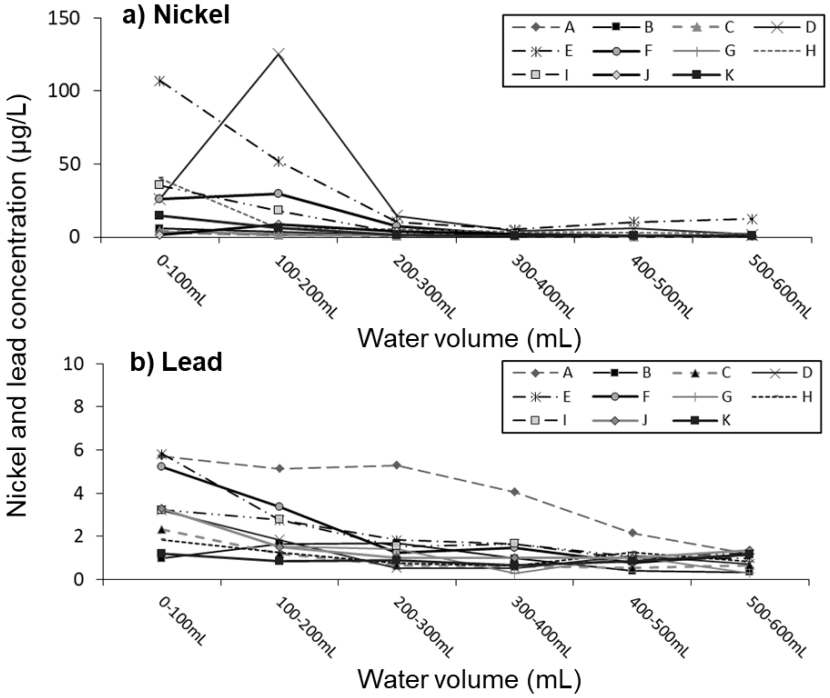

Fig. 3 Nickel (a) and lead (b) elution concentrations sequential flow from field taps

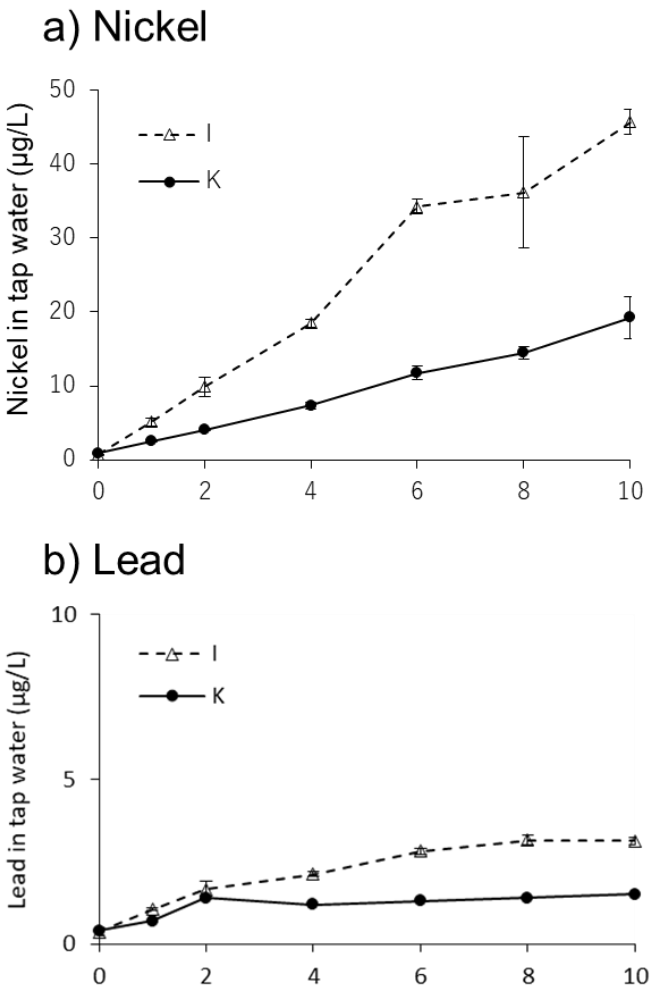

Fig. 4 Nickel (above) and lead (below) elution concentration in the first flow water from the taps after each retention period

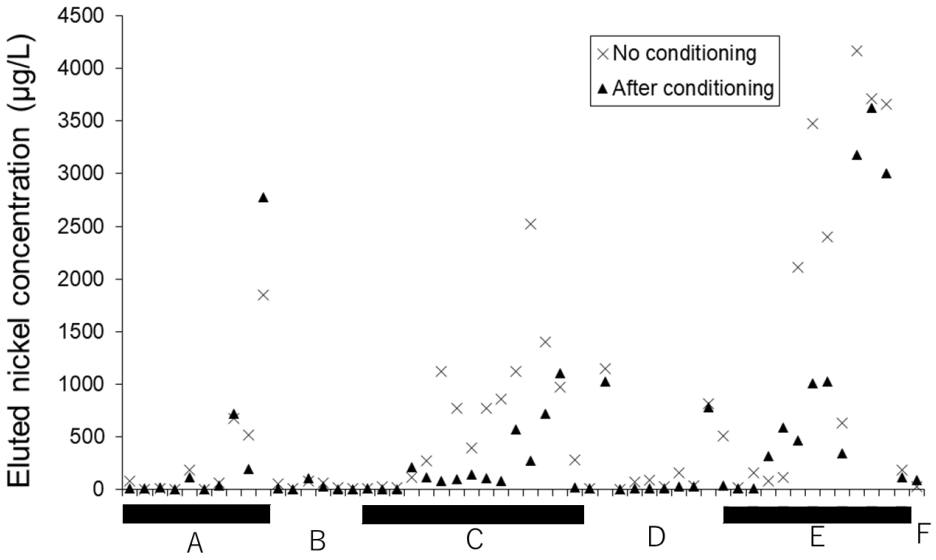

Fig. 5 Elution of nickel concentration by manufacturer

(A – F: company, details are shown in Table S1, not normalized with the inner volume)

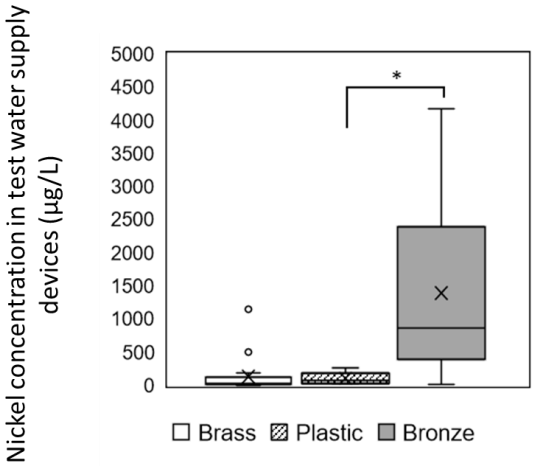

Fig. 6 Elution of nickel concentration by material

Supplementary Information

Table S1 Basic information on taps for continuous testing

	Type	Years	Manufacturer	Place
A	Single	14	TO	Veranda
B	Mix	14	TO	Kitchen
C	Mix	14	TO	Lavatory
D	Mix	0.5	Unknown	Lavatory
E	Mix	0.5	Unknown	Lavatory
F	Mix	0.5	Unknown	Kitchen
G	Mix	35	Unknown	Bath
H	Mix	35	Unknown	Kitchen
I	Mix	5	TO	Unknown
J	Mix	5	Unknown	Lavatory
K	Mix	5	Unknown	Bath

Fig. S1 Examples of taps
(left: single-lever mixer tap; middle: standard tap; right: double-handle mixer tap)

Fig. S2 Photograph of sequential sampling

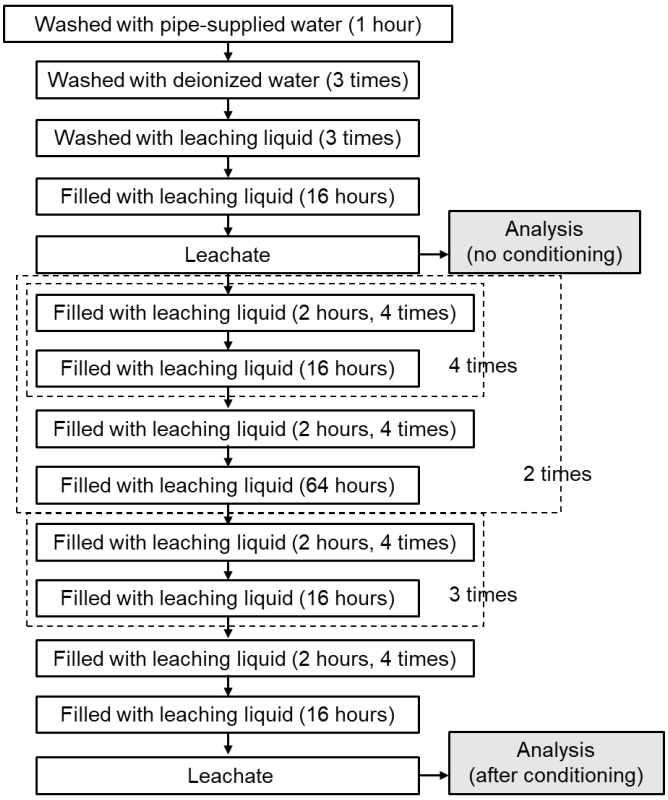

Fig. S3 Handling of the faucet and water for elution tests

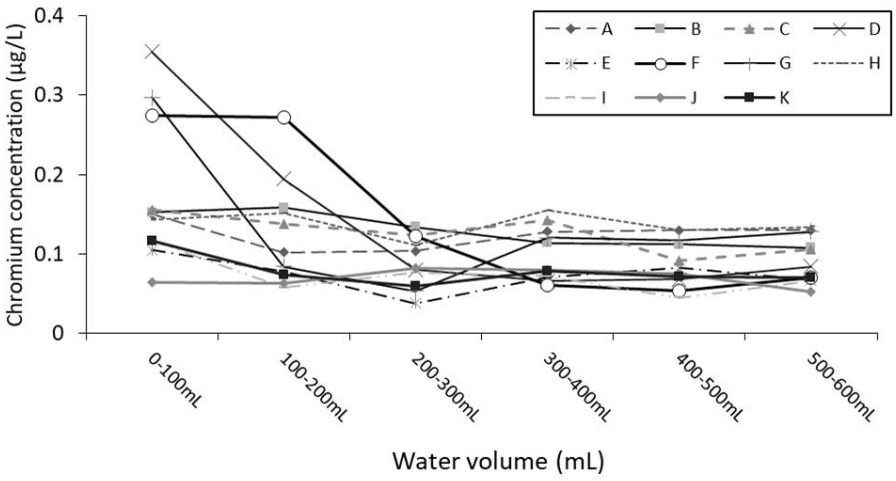

Fig. S4 Chromium elution concentration sequential flow from field taps

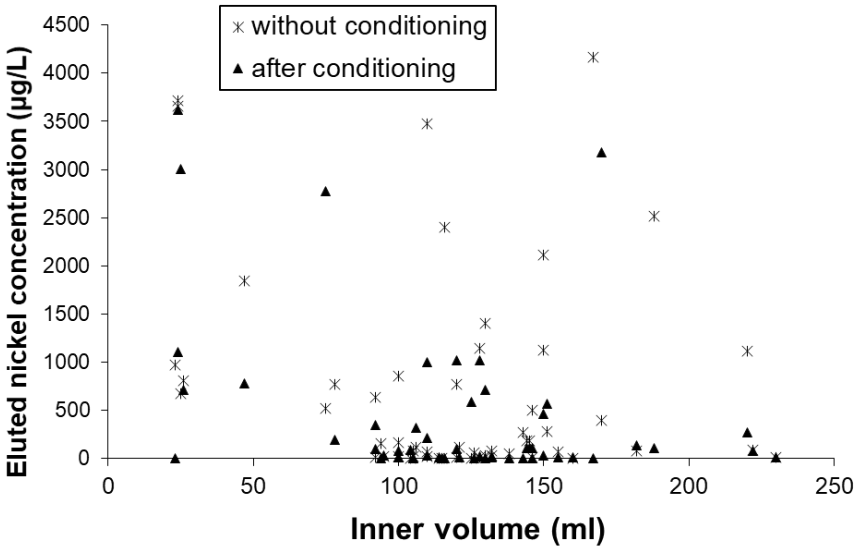

Fig. S5 Nickel concentration by inner volumes of faucets

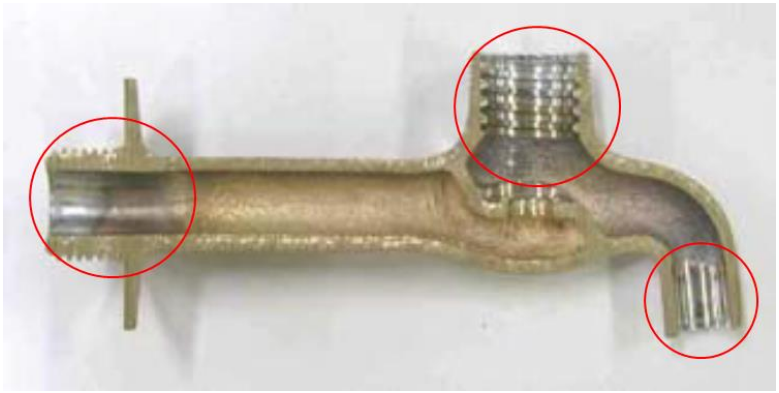

Fig. S6 Cross-section of a sample faucet with a nickel coating intruding inside

Red circles indicate nickel coating intruding inside.
Ref: Japan manufacturer's association

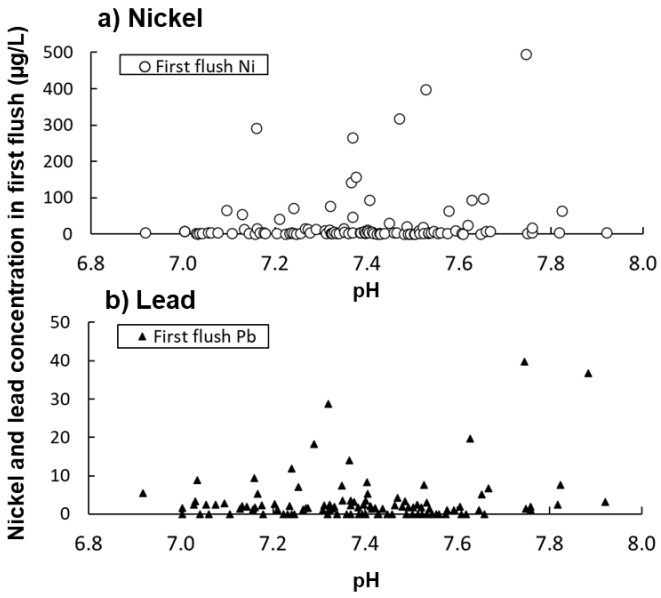

Fig. S7 Nickel and lead first flush concentrations and pH of the field survey taps

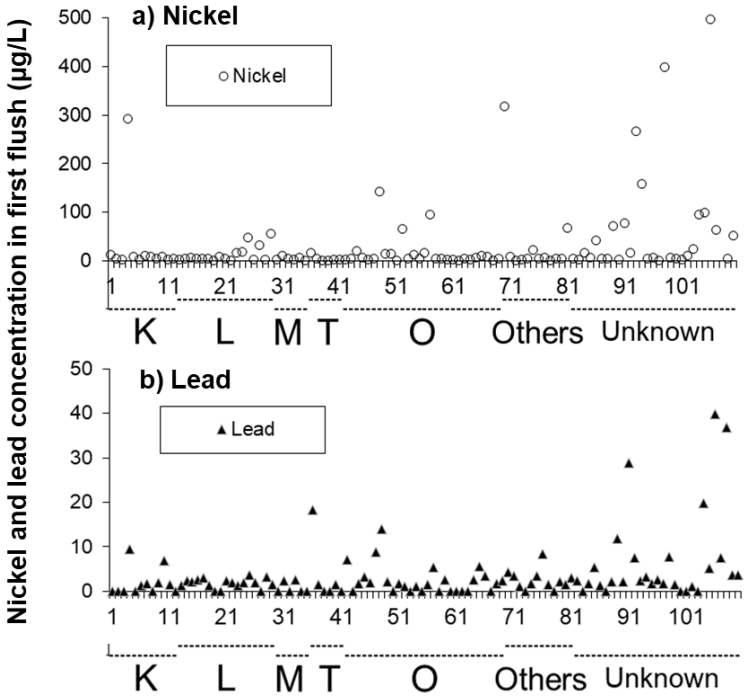

Fig. S8 Nickel and lead elution concentrations classified by manufacturer

of the field survey taps

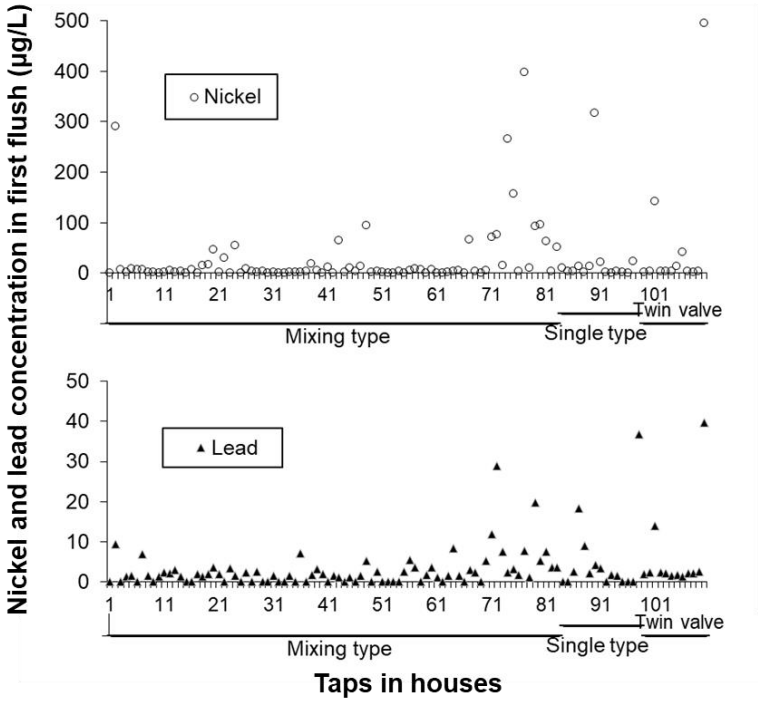

Fig. S9 Nickel and lead first flush concentrations according to the type of field survey taps

Credit author statement

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