To investigate potential health risks associated with exposure to metals from an abandoned metal mine, the authors studied people living near an abandoned mine (n=102) and control groups (n=149). Levels of cadmium, copper, arsenic, lead, and zinc were measured in the air, soil, drinking water, and agricultural products. To assess individual exposure, biomarkers of each metal in blood and urine were measured. β2-microglobulin, α1-microglobulin, and N-acetyl-beta-glucosaminidase and bone mineral density were measured. Surface soil in the study area showed 2-10 times higher levels of metals compared to that of the control area. Metal concentrations in the groundwater and air did not show any notable differences between groups. Mean concentrations of cadmium and copper in rice and barley from the study area were significantly higher than those of the control area (p<0.05). Geometric means of blood and urine cadmium in the study area were 2.9 μg/L and 1.5 μg/g Cr, respectively, significantly higher than those in the control area (p<0.05). There were no differences in the levels of urinary markers of early kidney dysfunction and bone mineral density. The authors conclude that the residents near the abandoned mine were exposed to higher levels of metals through various routes.

**Key Words**: Abandoned Metal Mine; Health Risk; Biological Markers; Kidney Dysfunction

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**INTRODUCTION**

Mining gives rise to soil erosion and environmental contamination by generating waste during the extraction, beneficiation, and processing of minerals. After closure, mines can still impact the environment by contaminating air, water, soil, and wetland sediments from the scattered tailings, as well as pollution of groundwater by discharged leachate, unless the proper remediation is conducted. Heavy metal contamination of agricultural soils and crops surrounding the mining areas is a serious environmental problem in many countries (1-5).

Coal and metal mining were actively developed in the Republic of Korea from the early 20th century, but most mines were closed in the 1970s due to poor productivity and exhaustion of ore reserves (4). By the year 2000, 906 abandoned metal mines, such as gold, copper (Cu), lead (Pb), and zinc (Zn), were scattered across the nation; most of these mines have been left without any management (5). Accordingly, large amounts of mine waste, such as abandoned structures and equipment, highwalls, open pits, mining dump tailings, lack of drainage control, acid water, and toxic materials, were left in unsafe and unhealthy conditions (6), increasing the potential health risk to the residents living near the mines. Heavy metal contamination of soil, water, and crops, and their health impact on residents, is a persistent social issue, and several studies have identified health risks of residents living near abandoned mines (7, 8).
To control the hazard posed by these abandoned metal mines, the Korean government regularly surveys the state of soil pollution near the mines and has constructed protective facilities. However, 8% of the abandoned mine sites have been remediated approximately, leaving many residents exposed to potential environmental hazards (4).

The lack of reliable information on the environmental pollution and health impact related to contamination of the abandoned site drew attention to the need for a community health study. The background of this investigation was a report suspecting a cadmium (Cd)-related health symptoms among the residents near an abandoned Cu mine located at the southern coast of Gyeongsangnam-do by a non-governmental organization (NGO). In this area, there were three abandoned Cu mines. The incidences of so called “itai-itai”-like symptom among the residents were hinted in mass-media, which raised nationwide public concerns. This investigation was initiated to understand the adverse health effects on heavy metals among the residents who lived near an abandoned metal mine.

In this study, the author investigated heavy metal concentrations in vicinity of abandoned mine and its health impact by conducting environmental measurements, biological monitoring, and health examinations on the residents.

MATERIALS AND METHODS

Study area and subjects

The abandoned mine is located in Byeongsan-ri, Goseong-gun, Gyeongsangnam-do, a small village in the southern part of Korean Peninsula (Fig. 1). The mine is located up-stream of the Byeongsan-ri operated from 1967 to early 1982, and was one of the sources of a small stream running through the village located 1.5 km downstream. Leachate of about 30 tons per day from one of the two openings flows into the stream across a small reservoir located at the foot of a mine tailings dam 300 meters downstream from the opening with 75,000 m$^3$ of storage. The residents of the village used the water from the polluted stream for drinking and irrigating their rice fields until they changed water sources in 1995. In addition, waste tailings had been stored without any treatment at the smelter near the mine until the government started a remediation project to prevent further contamination by the mine tailings and leachate in 1997. Environmental monitoring conducted by the Ministry of Environment and an NGO since the year 2000 revealed a high level of Cu, arsenic (As), Pb, and/or Cd in the soil, sediment of the bottom of the stream, leachate, and crops.

The village near the mine was selected as an exposed area, and two villages that showed similar socio-economic features with the exposed area were selected as control areas. Even though two control villages belong to the same county as the exposed area, they are more than 10 km away from the abandoned mine.

For the baseline survey, we visited all the households in the study areas to collect demographic information and to encourage people to participate in this study. We conducted health examinations for 102 participants from the exposed area, which amounts to 48.4% of the total population. From the two control areas, 149 participants were examined, which are 48.7% and 28.7% of the total population of each area.

Questionnaire

To characterize health status and concerns, we used two types of questionnaires, household and individual questionnaires. The household questionnaire was collected by door-to-door interviews. Subjects were required to fill out individual questionnaires when they received a health exam. Those who were illiterate or had difficulty in completing the questionnaires were aided by trained interviewers, six medical students. The household questionnaire elicited information about household information, place of residence, and past medical history. The individual questionnaire elicited information about demographic characteristics, education, job, tobacco and alcohol consumption, environmental exposure characteristics, and type of drinking water.

Environmental measurements

Environmental exposure to heavy metals was evaluated by collecting and analyzing environmental media samples (air, drinking water, and soil), with Cd, Pb, Cu, As, and Zn as target metals. These heavy metals were chosen as contaminants of potential concern based on other survey results in
Korea (9).

For the sampling, we have divided the study area into four districts by the level of contamination, based on the previous studies. Site of the sampling were evenly distributed over the study districts. However, oversampling was done at the district containing the major contamination source to ensure the estimation of the concentration gradient.

Forty surface soil samples were collected from residential areas and road sides of the study area, and 11 surface soil samples were collected from the control areas. Samples were analyzed by the method of Environmental Protection Agency 3050B and 6010B (10). Nine groundwater wells were sampled from the study area, and 3 samples from the control areas. For air samples, sites (three locations from the study area and two control sites) were chosen where the influence of traffic was negligible. PM$_{10}$ and total suspended particulates (TSP) were analyzed. From the PM$_{10}$ samples, heavy metal contents were analyzed. Samples were quantified using an inductively coupled plasma (ICP).

Rice samples were collected from 73 households in the exposed area and 24 households from the two control areas. Samples of barley, bean, sesame leaves, pepper, sweet potato stems, and oysters were also collected from 10 to 14 households. Samples were analyzed using an inductively coupled plasma mass spectrometry (ICP-MS) and atomic absorption spectrophotometer (AAS).

Biological monitoring

Exposure to heavy metals was evaluated by measuring Cd, Pb, Cu, and Zn in the blood and Cd in the urine. Blood samples were collected into EDTA tubes and stored at 4°C. Urine samples were collected into a urine bottle that was washed with HNO$_3$ prior to use. Measurement of heavy metal concentrations was conducted in a commercial laboratory with quality control certification from the Korea Occupational Safety and Health Agency. Whole blood samples were diluted in a mixed matrix modifier containing Triton X-100, nitric acid, and dibasic ammonium phosphate. The samples were analyzed on a flameless Zeeman atomic absorption spectrophotometer (Varian SpectrAA 800/GTA-100, Palo Alto, CA, U.S.A.).

All samples were analyzed in duplicate (and triplicate wherever possible). For blinding, biological samples were labeled with a random number generated independently from the original identifier or study group.

Health examination

Venous blood was drawn for the measurement of hemoglobin, total cholesterol, serum aspartate aminotransferase (AST), alanine aminotransferase (ALT), and serum iron and ferritin.

Spot urine samples were collected for Cd-induced kidney damage. Urine samples were adjusted to a pH and assayed using an available kit for the measurement of blood urea nitrogen (BUN), creatinine (Cr), uric acid, calcium, phosphorus, β$_2$-microglobulin (β$_2$-MG), θ$_1$-microglobulin (θ$_1$-MG), and N-acetyl-beta-glucosaminidase (NAG).

Bone mineral density was measured in each subject by peripheral dual energy radiography absorptiometry (p-DEXA, EXA-3000, OsteoSys Co., Seoul, Korea) at the distal end of the radius and calcaneus.

Statistical analysis

Homogeneity between groups was checked by the Student's t-test (age, height, weight, and years of residence) and the Chi-square test (gender, education, smoking, alcohol, drinking water). Descriptive statistics were used to analyze metal levels in environmental media and agricultural products, the blood and urine metal levels, and general health status. Statistical analysis of the two groups (the exposed and controls) was performed using the t-test and Mann-Whitney test. The level of significance was set at $p<0.05$. All analyses were performed with an SPSS statistical package (version 12.0) (SPSS Inc., Chicago, IL, U.S.A.).

Table 1. General characteristics of the subjects in the study and control area

|                        | Exposed (n=102) | Controls (n=149) | p value |
|------------------------|----------------|-----------------|---------|
| Age (yr)               | 62.5±13.7      | 61.7±14.2       | 0.495   |
| Height (cm)            | 157.1±9.6      | 158.2±9.7       | 0.397   |
| Weight (kg)            | 56.7±11.1      | 57.5±10.5       | 0.542   |
| Gender (%)             |                |                 | 0.698   |
| Male                   | 45 (44.1)      | 61 (40.9)       |         |
| Female                 | 57 (55.9)      | 88 (59.1)       |         |
| Education (%)          |                |                 | 0.656   |
| No education           | 37 (36.6)      | 83 (33.6)       |         |
| Primary school         | 34 (33.7)      | 81 (32.8)       |         |
| Middle school          | 15 (14.9)      | 39 (15.8)       |         |
| High school            | 13 (12.9)      | 40 (16.2)       |         |
| College                | 2 (2.0)        | 4 (1.6)         |         |
| Smoking (%)            |                |                 | 0.311   |
| Yes                    | 28 (27.7)      | 46 (31.5)       |         |
| No                     | 73 (72.3)      | 100 (68.5)      |         |
| Alcohol (%)            |                |                 | 0.189   |
| Yes                    | 35 (34.7)      | 111 (44.8)      |         |
| No                     | 66 (65.3)      | 137 (55.2)      |         |
| Job (%)                |                |                 | 0.221   |
| Agriculture/fishery    | 73 (72.3)      | 95 (65.5)       |         |
| Others                 | 28 (27.7)      | 50 (34.5)       |         |
| Drinking water (%)     |                |                 | 0.212   |
| Simple piped water     | 54 (66.7)      | 79 (62.2)       |         |
| Well in the household  | 13 (16.0)      | 43 (33.9)       |         |
| Mixed usage            | 14 (17.3)      | 5 (3.9)         |         |
| Duration of residence (yr) | 44.2±21.3    | 40.0±23.0       | 0.193   |
RESULTS

There were no significant differences in the general or demographic characteristics of the exposed and control populations (Table 1). The Cd, Cu, As, Pb, and Zn levels in surface soil of the exposed area were significantly higher compared to the control area (*p<0.05; Table 2). In drinking water, only Zn was higher in the exposed area than the control area. Airborne PM10 levels in the exposed area were significantly higher than the control area (*p<0.05).

The spatial distribution of Cd in soil was analyzed using the geometric information system (GIS) method. Two hot spots of high Cd concentration were identified on the map, which correspond to the mine and the tailing site (Fig. 2). Cd levels in rice and barley from the exposed area were significantly higher than the control area (*p<0.05; Table 3). In particular, rice and barely samples collected from locations near the metal mines or from the area influenced by mine drainage showed significantly higher Cd levels compared to those from the control area (*p<0.05).

Table 4 shows the results of heavy metal concentrations in blood or urine by residential area. Blood and urine Cd concentrations in the exposed area were significantly higher than in the control area (*p<0.05). The results showed that people living in the area near the metal mines have been exposed to Cd. However, means of blood pressure, cholesterol, and BUN/creatinine were not significantly different between the groups.

Table 3. Heavy metal levels in agricultural products (unit: mg/kg)

| Group          | Cd             | Cu             | As              | Pb               | Zn*            |
|----------------|----------------|----------------|-----------------|------------------|---------------|
| Rice           |                |                |                 |                  |               |
| Exposed        | 45 (45) ± 0.151| 24 (24) ± 0.521| 5 (5) ± 0.186   | 5 (5) ± 0.015    | 5 (5) ± 0.015 |
| p value        | 0.026          | 0.013          | 0.212           | 0.040            | 0.040         |
| Control        | 45 (24) ± 0.151| 24 (24) ± 0.521| 5 (5) ± 0.186   | 5 (5) ± 0.015    | 5 (5) ± 0.015 |
| p value        | 0.026          | 0.013          | 0.212           | 0.040            | 0.040         |
| Barley         |                |                |                 |                  |               |
| Exposed        | 7 (7) ± 0.005  | 6 (6) ± 0.011  | 6 (6) ± 0.011   | 6 (6) ± 0.011    | 6 (6) ± 0.011 |
| p value        | 0.001          | 0.011          | 0.001           | 0.011            | 0.011         |
| Control        | 7 (7) ± 0.005  | 6 (6) ± 0.011  | 6 (6) ± 0.011   | 6 (6) ± 0.011    | 6 (6) ± 0.011 |
| p value        | 0.001          | 0.011          | 0.001           | 0.011            | 0.011         |
| Bean           |                |                |                 |                  |               |
| Exposed        | 10 (10) ± 0.015| 5 (5) ± 0.186  | 5 (5) ± 0.212   | 5 (5) ± 0.015    | 5 (5) ± 0.015 |
| p value        | 0.040          | 0.040          | 0.040           | 0.040            | 0.040         |
| Control        | 10 (10) ± 0.015| 5 (5) ± 0.186  | 5 (5) ± 0.212   | 5 (5) ± 0.015    | 5 (5) ± 0.015 |
| p value        | 0.040          | 0.040          | 0.040           | 0.040            | 0.040         |
| Sesame leaves  |                |                |                 |                  |               |
| Exposed        | 10 (10) ± 0.015| 9 (9) ± 0.176  | 9 (9) ± 0.364   | 9 (9) ± 0.017    | 9 (9) ± 0.017 |
| p value        | 0.001          | 0.001          | 0.001           | 0.001            | 0.001         |
| Control        | 10 (10) ± 0.015| 9 (9) ± 0.176  | 9 (9) ± 0.364   | 9 (9) ± 0.017    | 9 (9) ± 0.017 |
| p value        | 0.001          | 0.001          | 0.001           | 0.001            | 0.001         |
| Hot pepper     |                |                |                 |                  |               |
| Exposed        | 15 (15) ± 0.015| 15 (15) ± 0.176| 15 (15) ± 0.248 | 15 (15) ± 0.004  | 15 (15) ± 0.004 |
| p value        | 0.011          | 0.011          | 0.011           | 0.011            | 0.011         |
| Control        | 15 (15) ± 0.015| 15 (15) ± 0.176| 15 (15) ± 0.248 | 15 (15) ± 0.004  | 15 (15) ± 0.004 |
| p value        | 0.011          | 0.011          | 0.011           | 0.011            | 0.011         |
| Stem of sweet potato | | | | | |
| Exposed        | 8 (8) ± 0.004  | 10 (10) ± 0.155| 10 (10) ± 0.012 | 10 (10) ± 0.007  | 10 (10) ± 0.007 |
| p value        | 0.003          | 0.003          | 0.003           | 0.003            | 0.003         |
| Control        | 8 (8) ± 0.004  | 10 (10) ± 0.155| 10 (10) ± 0.012 | 10 (10) ± 0.007  | 10 (10) ± 0.007 |
| p value        | 0.003          | 0.003          | 0.003           | 0.003            | 0.003         |
| Oyster         |                |                |                 |                  |               |
| Exposed        | 14 (14) ± 0.224| 14 (14) ± 0.212| 15 (15) ± 0.156 | 14 (14) ± 0.156  | 14 (14) ± 0.156 |
| p value        | 0.014          | 0.014          | 0.014           | 0.014            | 0.014         |
| Control        | 14 (14) ± 0.224| 14 (14) ± 0.212| 15 (15) ± 0.156 | 14 (14) ± 0.156  | 14 (14) ± 0.156 |
| p value        | 0.014          | 0.014          | 0.014           | 0.014            | 0.014         |

Table 2. Heavy metal levels in the soil, drinking water, and air

|         | Soil (mg/kg) | Drinking water (mg/L) | Air (μg/m³) |
|---------|--------------|-----------------------|-------------|
|         | No. of samples | Mean ± SD | No. of samples | Mean ± SD | No. of samples | Mean ± SD |
|         |               |            |               |            |               |            |
| Cd      | 11 (11)       | 0.355 ± 0.263  | 11 (11)       | 0.182 ± 0.136 |
| Cu      | 11 (11)       | 268.2 ± 599.8   | 11 (11)       | 66.37 ± 128.3 |
| As      | 11 (11)       | 22.53 ± 36.44   | 11 (11)       | 7.296 ± 3.973 |
| Pb      | 11 (11)       | 2.176 ± 4.37    | 11 (11)       | 23.33 ± 12.66 |
| Zn      | 11 (11)       | 29.60 ± 58.8    | 11 (11)       | 1.92 ± 1.76   |

* Number of samples (number of samples above detection limit); †, Not applicable; ‡, Maximum level.
*, p<0.05 by Student’s t-test.
Cadmium in soil shows two hot spots by concentration gradient graphics. One, the strongest, coincides with the entrance of the abandoned mine (A), and the other, smaller one, located at the mine tailing dam (B). Thus, the source of cadmium in the soil in the area is the abandoned mine. Unit, mg/kg; *, abandoned mine; ◆, household.

Table 4. Heavy metal levels in blood and urine (geometric mean, Standard deviation [range])

|        | Exposed (n=102) | Controls (n=149) | p-value |
|--------|-----------------|------------------|---------|
| Cd_B (μg/L) | 2.92 ± 1.67 (0.70-14.55) | 2.05 ± 0.98 (0.26-8.87) | 0.000 |
| Cd_U (μg/g Cr) | 1.53 ± 0.61 (0.10-11.63) | 1.20 ± 0.71 (0.08-14.71) | 0.012 |
| Cu_U (μg/L) | 14.03 ± 0.02 (0.40-79.34) | 14.31 ± 0.30 (1.38-76.25) | 0.700 |
| Pb_B (μg/dL) | 2.37 ± 1.75 (0.80-16.16) | 2.46 ± 2.63 (0.53-9.28) | 0.439 |
| Zn_U (μg/L) | 309.04 ± 37.64 (2.49-2,442.21) | 313.75 ± 363.31 (14.35-2,070.97) | 0.850 |

Cd_B, cadmium in blood; Cd_U, cadmium in urine; Cu_U, copper in urine; Pb_B, lead in blood; Zn_U, zinc in urine.

Cr were not different between the two groups (Table 5). The prevalences of hypertension in the exposed area and the control area were 40.8% and 47.0%, respectively, showing no significant difference (p>0.05). Health effects indicators, including renal dysfunction indices and bone mineral density, were not significantly different between the two groups (p>0.05) (Table 6).

Table 5. General health status of the subjects in the study and the control area (mean±SD)

|          | Exposed (n=102) | Controls (n=149) | p-value |
|----------|-----------------|------------------|---------|
| Systolic BP (mmHg) | 128.9±18.6 | 130.7±16.1 | 0.411 |
| Diastolic BP (mmHg) | 80.8±11.0 | 83.8±11.0 | 0.037 |
| BMI      | 22.8±3.2 | 22.9±3.2 | 0.861 |
| Hemoglobin (g/dL) | 13.3±1.6 | 13.2±1.5 | 0.604 |
| Total cholesterol (mg/dL) | 192.6±31.7 | 189.7±37.5 | 0.524 |
| AST (IU/L) | 27.4±25.7 | 24.8±16.7 | 0.296 |
| ALT (IU/L) | 24.5±27.5 | 22.6±19.2 | 0.520 |
| Urea nitrogen (mg/dL) | 18.6±4.8 | 17.2±4.8 | 0.485 |
| Creatinine (mg/dL) | 1.0±0.2 | 1.0±0.2 | 0.510 |
| Uric acid (mg/dL) | 5.1±1.6 | 5.4±1.7 | 0.589 |
| Calcium (mg/dL) | 9.2±0.4 | 9.3±0.4 | 0.171 |
| Phosphorus (mg/dL) | 3.8±0.6 | 3.9±0.6 | 0.749 |
| Serum iron (μg/L) | 98.0±37.4 | 101.6±43.9 | 0.500 |
| Ferritin (μg/L) | 112.1±193.9 | 101.5±105.6 | 0.564 |

BP, blood pressure; BMI, body mass index; AST, serum aspartate aminotransferase; ALT, serum alanine aminotransferase.

Table 6. Health outcomes related to cadmium exposure in the study and reference area (mean±SD)

|        | Exposed (n=102) | Controls (n=149) | p-value |
|--------|-----------------|------------------|---------|
| Renal damage indicators β-MG (μg/g Cr) | 267.7±149.9 | 240.4±168.9 | 0.284 |
| α-MG (μg/g Cr) | 4.12±3.03 | 4.36±4.26 | 0.733 |
| NAG (U/g Cr) | 5.82±3.96 | 5.38±4.54 | 0.437 |
| Osteoporosis indicator Calcaneal p-DX A* | -1.49±1.35 | -1.38±1.42 | 0.300 |
| β-MG, β-microglobulin; α-MG, α-microglobulin; NAG, N-acetyl-beta-glucosaminidase; p-DX A, Peripheral dual energy x-ray absorptiometry in calcaneus. *T-score.

DISCUSSION

We examined whether an abandoned mine had adverse effects on the environment and people living nearby. Blood and urinary Cd levels of people who lived near the mine were significantly higher than those of control site residents. Levels of blood Cd in the exposed area were also higher than the general Korean population in the 2005 Korean National Health and Nutrition Examination Survey, in which Cd levels were 1.52 μg/L among all age groups and 1.65 μg/L among people 60 yr or older (5). However, in our study, high Cd levels did not correlate with changes in bone mineral density or tubular damage in the kidney. These elevated Cd levels did not correlate with changes in bone mineral density or detectable health effects. Our result indicate that the residents in the study area are exposed to Cd originated from the abandoned mine, but the levels are not sufficient to cause detectable health effects. However, considering that European studies have shown signs of Cd-induced kidney dysfunction in the general population at
urinary Cd levels of 2-3 μg/g Cr (11, 12), we need to follow up the study population for possible adverse health effects.

Rice and barely samples from locations near the mines or influenced by mine drainage showed significantly higher Cd levels than the control area. The fact that the level of Cu showed a similar pattern with Cd supports the hypothesis that higher levels of Cd in rice and barley were derived from the abandoned Cu mine. Cd levels in rice and barley may have been much higher than the present levels before the remediation project was completed. Food is the most significant source of Cd exposure in the general, non-smoking population (13). Because rice is the main source of food for residents, people in the area probably had been exposed to Cd through eating rice. In case of Cu, levels in rice and barley from the exposed area were higher than those of the control area. However, they may not be sufficient to affect the concentrations of blood Cu. The level of copper in the agricultural products of the exposed area was less than 3 percent of the provisional tolerable daily intake level.

Cd levels in the air were below the detection limit in all areas, probably because the waste tailing was covered with soil after remediation work. However, we can exclude the possibility that residents were exposed to elevated Cd levels before that. In fact, resident testimony indicated that they had problems with waste site dust in the past, suggesting greater inhalation exposure before remediation.

People in the exposed area drank water either from water supply facility, which is sourced far from the mine, or from the private wells. The level of Cd in the water people drank at the time of our investigation was below the detection limit. The levels of other heavy metals such as Cu, Zn, and Pb were also negligible. Although it is not likely that the water people drank at time of the investigation were contaminated by heavy metals, people may have consumed polluted water before water service started 10 yr ago. Though the levels of arsenic in the wells were higher in the exposed area than in the control area, we were not able to assess the health implications due to lack of available biomonitoring data.

There have been many studies on the health risks of abandoned mines, usually focused on environmental mediums (e.g., air, water, or soil) (14-18). The strength of our study is its holistic approach, allowing comprehensive evaluation of the effects of heavy metal exposure from the source to the health outcomes via levels of environmental media and body burden. Through this approach, we determined how people living near the mine had accumulated a higher level of Cd, which originated from the abandoned Cu mine.

However, not all measurements contributed equally to the exposure assessment. We could not detect Cd in the air or water, implying that current levels of environmental heavy metals may have limited predictive potential for previous exposure. In contrast, urinary Cd was a good indicator for exposure to Cd over the past several decades.

We suggest that biologic monitoring be considered a first choice to monitor the possible health effects of residents living near abandoned mines. Further studies using more sensitive measurement of exposure and health effects would reveal the health impact of heavy metal exposure from abandoned mines.

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