

## Research Note

# Heavy Metals in Spring and Bottled Drinking Waters of Sibylline Mountains National Park (Central Italy)

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## ABSTRACT

Heavy metal concentrations (cadmium, lead, and copper) in spring, tap, and bottled waters of the Sibylline Mountains National Park (central Italy) were investigated using square wave anodic stripping voltammetry from 2004 to 2011. The mean ( $\pm$ SD) concentrations detected ( $1.3 \pm 0.4$  ng L<sup>-1</sup> cadmium,  $14 \pm 6$  ng L<sup>-1</sup> lead, and  $0.16 \pm 0.10$   $\mu$ g L<sup>-1</sup> copper) were below the limits stipulated by Italian and European legislation for drinking and natural mineral water. In the three studied areas of the park (Mount Bove north, Mount Bove south, and springs of River Nera) with very few exceptions, both mineral waters bottled in the area and aqueduct waters from public fountains had approximately the same metal concentrations as did the spring waters from which they were derived. Conversely, substantially higher metal concentrations were found at some sites in private houses, which may be due to release of metals from old metal pipes. At the time of this study, waters of Sibylline Mountains National Park were of good quality, and no influence of the bottling process on heavy metal concentrations was found.

**Key words:** Cadmium; Copper; Lead; Mineral water; Spring water; Tap water

Human consumption of mineral water has increased in recent years because of problems related to water resources in some countries (23) and for reasons linked to flavor and health benefits. Sales of mineral water have rapidly increased in the last two decades, and in Europe alone, 2,000 mineral water brands are available. Although many people consider mineral water safer than tap water, the concentrations of toxic natural elements or heavy metals can sometimes exceed regulatory limits (39). The presence of these contaminants could be related to soil properties (14, 18, 41), and minor elements such as cadmium (Cd), lead (Pb), copper (Cu), iron, and zinc can enter water via pipeline corrosion (50) or leaching from bottles (38, 47, 48). Many materials (e.g., brass valves and leaded solders (11, 46, 56)) may be sources of these elements, and factors such as pH and alkalinity can influence the corrosion and release of these elements from the water distribution system (29).

Few studies have dealt with trace elements in drinking and natural mineral waters (8, 9, 12, 13), and no studies have been conducted to compare heavy metal concentrations in ground water and drinking water (tap and bottled) to ensure the safety of water after bottling and distribution.

The main objective of this work was the characterization of the heavy metal concentrations in drinking water collected from the Sibylline Mountains National Park (Marche Region, central Italy). In particular, our goal was to determine the baseline concentrations of Cd, Pb, and Cu in drinking water from one of the most important source

areas of spring waters of the Marche region and to assess any changes in the spring water between the spring and bottled water, aqueduct water, or tap water. The three elements investigated are widely used in the pipe materials of water distribution systems. From a public health perspective, this study was conducted to provide consumers with a better understanding of the properties of drinking water, enabling consumers to make informed choices at the time of purchase.

## MATERIALS AND METHODS

**Study area.** The study was carried out in the Sibylline Mountains National Park in the Apennine Mountains of central Italy. Twenty-three water samples (spring, mineral, and tap) were collected in three areas of the park from 2004 to 2011: Mount Bove north (12 samples), Mount Bove south (5 samples), and the springs of the River Nera (6 samples).

**Sampling and sample storage and treatments.** Water samples were collected following national procedures (36) directly in polyethylene sampling bottles that were previously decontaminated as described elsewhere (2, 3, 26, 43, 44). All samples were immediately placed in a refrigerated bag and rapidly transported for analysis (within 2 h).

**On-site measurements.** Temperature and electrical conductivity (EC) at 20°C were measured at the sampling sites immediately after collection using a handheld conductivity meter (model 30, YSI, Yellow Springs, OH) as described in the Supplemental Material.

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### Laboratory, apparatus, and decontamination procedures.

A class 6 clean room (International Organization for Standardization, Geneva, Switzerland) with class 5 areas under laminar flow cabinets was used for material decontamination procedures, sample processing, and metal analyses as reported elsewhere (6, 7, 45, 53).

**Reagents.** All solutions were prepared using Milli-Q water (Millipore, Bedford, MA); ultrapure and superpure acids were from Romil (Cambridge, UK). Others materials were obtained from Carlo Erba (Milan, Italy): analytical grade HNO<sub>3</sub>; atomic absorption spectrometry standards of Cd (II), Pb (II), and Cu (II); EDTA; analytical grade NaCl, NaF, KI; total ionic strength adjustment buffer III; and LGG09041910. Eriochrome Black T was obtained from Hamilton (Bonaduz, Switzerland), and sodium standard solution and ionic strength adjusters were purchased from Thermo Orion Corp. (Beverly, MA)

**Chemical physical analysis.** Total dissolved solids (TDS), total water hardness, and concentrations of major ions (Na<sup>+</sup>, Cl<sup>-</sup>, F<sup>-</sup>, I<sup>-</sup>, and NO<sub>3</sub><sup>-</sup>) were measured in the laboratory as described in the Supplemental Material.

**Metal analyses.** Metal concentrations were determined by square wave anodic stripping voltammetry (SWASV) as described elsewhere (1, 2, 25) using a 746 VA trace analyzer and a 747 VA stand (Metrohm, Riverview, FL). The main voltammetric parameters were set to optimize the procedure for the analysis of mineral water: deposition potentials of -1,050 (Cd) and -975 (Pb and Cu) mV versus Ag/AgCl, 3 mol/L KCl; deposition time of 15 (Pb and Cu) to 45 (Cd) min; SW amplitude of 25 mV; SW frequency of 100 Hz; step height of 8 mV; and step time of 100 ms.

**Laboratory blanks, analytical variability, and accuracy.** SWASV analysis of laboratory blanks (10 to 15 mL of ultrapure HCl, 0.036 mol L<sup>-1</sup>) verified that the metal concentration was below the limit of detection (LOD) of the method (52), i.e., approximately 0.2 ng L<sup>-1</sup> for Cd, Pb, and Cu (1-3). The analytical variability, estimated by the pooled standard deviation (SD<sub>pooled</sub>) (49) computed from the repeated measurements of the whole data set, was 19% for Cd, 15% for Pb, and 8% for Cu.

The accuracy of the analytical procedure was controlled by analyzing the certified reference material NASS-5 (35) for trace metals in seawater. Cd, Pb, and Cu concentrations were in agreement with the reference values within experimental errors. Mean (±SD) values (*n* = 8) were 27 ± 4 ng L<sup>-1</sup> Cd, 9 ± 2 ng L<sup>-1</sup> Pb, and 274 ± 41 ng L<sup>-1</sup> Cu versus certified values of 23 ± 3 ng L<sup>-1</sup> Cd, 8 ± 5 ng L<sup>-1</sup> Pb, and 297 ± 46 ng L<sup>-1</sup> Cu.

**Statistical analysis.** Data analyses were performed using Statgraphics (51) and Simca (42) packages. A descriptive analysis of the variables was carried out, and then a multivariate statistical analysis (principal component analysis) was used as a descriptive tool to visualize the data structure in a few dimensions, finding relationships between variables and area. Details are given in the Supplemental Material.

## RESULTS AND DISCUSSION

### Chemical and physical characteristics of water.

Chemical and physical data (temperature, EC, pH, TDS, Na<sup>+</sup>, Cl<sup>-</sup>, F<sup>-</sup>, I<sup>-</sup>, NO<sub>3</sub><sup>-</sup>) for all samples analyzed (in part reported previously (53)) are summarized in Supplemental

Table S1 for each of the three areas: Mount Bove north, Mount Bove south, and the springs of the River Nera.

According to Italian law (Table 1), the water of the Sibylline Mountains National Park have the following characteristics: oligomineral features (TDS of 50 to 500 mg L<sup>-1</sup>), medium hardness (8 to 12°f [80 to 120 ppm]), suitability for low sodium diets (Na < 20 mg L<sup>-1</sup>), and suitability for infant consumption (F<sup>-</sup> < 1.5 mg L<sup>-1</sup> and NO<sub>3</sub><sup>-</sup> < 10 mg L<sup>-1</sup>). Additional details are reported in the Supplemental Material.

**Heavy metals.** All results (in part reported previously (53)) are reported in Table 2.

**Heavy metals: Cd.** Cd concentrations were very low and similar in the three areas, with an overall mean (±SD) of 1.3 ± 0.4 ng L<sup>-1</sup> (relative standard deviation [RSD], 31%; range, 0.7 to 2.4 ng L<sup>-1</sup>), well below (about 2,000 and 3,500 times lower) the limits stipulated by major national and international institutions (Table 1). No significant differences were found between springs in the same area, and very similar values were detected across years.

Cd concentrations in some samples of aqueduct water from a rural house in Sorbo (Italy) were outliers: 4.4 ng L<sup>-1</sup> in 2009 and 2.0 ng L<sup>-1</sup> in 2011. These remarkable differences with respect to the mean values at this site (1.2 ± 0.3 ng L<sup>-1</sup>) could be ascribed to the old material in the distribution pipeline, which was probably made of asbestos cement, a material rich in Cd (21).

For Cd concentrations in bottled versus spring mineral water, a difference was observed between the Roana brand bottled mineral water (0.8 ng L<sup>-1</sup>) and the Panico spring water (1.7 ng L<sup>-1</sup>) (from which the Roana bottled water derives), with approximately twice the concentration in the latter. However, this difference was not significant (*P* > 0.05) because of the low overall Cd concentration (very close to the LOD of the technique), and the Cd concentrations were considered safe for consumers in both cases.

The opposite situation was found in the area of the River Nera, where we measured Cd at 1.1 ng L<sup>-1</sup> in the Uccelletto sample (from the main spring used for the Nerea brand bottled water) and 2.4 ng L<sup>-1</sup> in the Nerea bottled water sample. This increase in the Cd concentration in the bottled mineral water may be ascribed to the bottling process; however, the final concentration was still well below the legal limit (Table 1), which ensured safety for consumers.

Data obtained in this study were compared with published data (Table 3). For mineral waters, Cd concentration in the present study (mean, 1.6 ng L<sup>-1</sup>) was the same as those detected in other Italian and international samples (10, 37) but was 200- to 3,000-fold lower than that measured in Alabama (24). The same situation applies to tap water and spring water, for which our Cd data are in agreement with Italian and European values but are well below data reported for other parts of the world (27, 49).

TABLE 1. Maximum health-related limits from Italy, the European Union (EU), the EPA, and the FAO and guideline values from the WHO for drinking waters (i.e., intended for human consumption), spring waters, and natural mineral waters

Parameter	Water intended for human consumption (and spring water only for Italian law D.Lgs 176/11 (33)); Italian law D.Lgs 31/2001 (31), EU directive 98/83/EC (15)	Natural mineral water			
		Italian law D.Lgs 171/2011 (33); DM 29 Dec. 2003 (32) and 10 Feb. 2015 (34); EU directives 2003/40/EC (16) and 2009/54/EC (17)	FAO (19)	Drinking water	
			EPA (54, 55)	WHO (57)	
EC at 20°C ( $\mu\text{S}/\text{cm}$ )	2,500 <sup>a</sup>	ND <sup>b</sup>			
pH	6.5–9.5 <sup>a</sup>	ND		6.5–8.5 <sup>c</sup>	6.5–8.5
Cadmium ( $\mu\text{g L}^{-1}$ )	5.0 <sup>d</sup>	3.0	3	5	3 g.v. <sup>b</sup>
Chloride ( $\text{mg L}^{-1}$ )	250 <sup>a</sup>	ND			<250
Copper ( $\text{mg L}^{-1}$ )	2.0, <sup>d</sup> 1.0 <sup>a,e</sup>	1.0	1	1.3, 1.0 <sup>c</sup>	2
Fluorides ( $\text{mg L}^{-1}$ )	1.5 <sup>d</sup>	5.0 (1.5 <sup>f</sup> )	1, <sup>c</sup> 1.5 <sup>f</sup>	4.0, 2.0 <sup>g</sup>	1.5
Hardness ( $^{\circ}\text{f}$ )	15–50 <sup>a,h</sup>	ND			<200
Iodide ( $\text{mg L}^{-1}$ )		ND			
Lead ( $\mu\text{g L}^{-1}$ )	10 <sup>d</sup>	10	10	15	10
Nitrates ( $\text{mg L}^{-1}$ )	50 <sup>d</sup>	45 (10 <sup>f</sup> )	50	10	50
Sodium ( $\text{mg L}^{-1}$ )	200 <sup>a</sup>	ND (20 <sup>i</sup> )			
TDS at 180°C ( $\text{mg L}^{-1}$ )	1,500 <sup>a,j</sup>	ND		500 <sup>g</sup>	

<sup>a</sup> Indicator parameter.

<sup>b</sup> ND, not determined; g.v., guideline values.

<sup>c</sup> Secondary standard (only cosmetic or aesthetic effects).

<sup>d</sup> Chemical parameter.

<sup>e</sup> Value from Italian law is 1.0  $\text{mg L}^{-1}$ .

<sup>f</sup> Suitable in food for children (19, 34).

<sup>g</sup> Limit above which the label should include “contains fluoride.”

<sup>h</sup> Suggested values (only Italian law).

<sup>i</sup> Suitable for low-sodium diet (17, 33).

<sup>j</sup> Maximum suggested value (only Italian law).

**Heavy metals: Pb.** The mean Pb concentration in all analyzed waters (except those in Sorbo) was  $14 \pm 6 \text{ ng L}^{-1}$  (RSD, 43%; range, 6 to  $25 \text{ ng L}^{-1}$ ) (Table 3). These values were well below the limits stipulated by national and international authorities in both mineral and drinking waters of 10 and  $15 \mu\text{g L}^{-1}$ , respectively (Table 1).

Substantial differences were found for Pb distribution among the three areas, from Mount Bove south ( $7 \pm 2 \text{ ng L}^{-1}$ ), to River Nera springs ( $11 \pm 4 \text{ ng L}^{-1}$ ), to Mount Bove north ( $18 \pm 3 \text{ ng L}^{-1}$ ). There were no significant differences in the Pb concentration between springs of the same area, but very different values were recorded for aqueduct water. In the Mount Bove north area, Pb concentration was  $16 \pm 1 \text{ ng L}^{-1}$  (RSD, 6%) at the Frontignano public fountain and higher values,  $23 \pm 3 \text{ ng L}^{-1}$  (RSD, 13%), were found at the Ussita fountain. This difference is probably due to differences in water pipes; lead was commonly used in pipes in the past (21). Clear evidence of contamination from pipes was obtained from the Sorbo rural house water, where Pb concentration was 5- to 10-fold higher than those in spring waters in the same area. This house had old pipes presumably made of lead (e.g., in cement mortar materials), and leaching into the drinking water was a very likely possibility (22). This type of contamination reflects the problem of the “last mile” of water works, where the composition of the material in the water distribution system, the age of the distribution system, and the interaction time between water and material, such as Pb in solder, can influence the presence of many elements (12).

In a comparison of Roana bottled mineral water with the Panico spring water from which the bottled water was derived, no significant differences were found for Pb, which had the same concentration ( $16 \text{ ng L}^{-1}$ ) in both samples. Thus, we assumed that the bottling procedure at the factory was free of contamination by this metal.

Nerea bottled mineral water originates from the Uccelletto spring. In this case, Pb concentration in the bottled water ( $11 \text{ ng L}^{-1}$ ) was twice that in the corresponding spring sample ( $7 \text{ ng L}^{-1}$ ), similar to the situation found for Cd. For both metals, a remarkable increase was revealed in the passage from spring to bottle, suggesting a possible effect of the bottling procedure on Cd and Pb concentrations. However, the Pb concentration was well below the limits stipulated by government regulations, supporting the good quality of the bottled drinking water.

In a comparison of the Pb concentration of Sibylline water with that previously published, our results are similar to measurements for Italian and European mineral and tap waters (10, 13, 37) but 5- to 10,000-fold lower than the Pb concentrations reported for other countries (24, 27, 39). For spring waters, only limited published data are available; however, our results for Pb were five to eight times lower than those reported for the Mount Reventino area (4) (Table 3).

**Heavy metals: Cu.** Cu concentrations were higher than those of Cd and Pb in the Sibylline waters, at  $0.16 \pm 0.10 \mu\text{g L}^{-1}$  (RSD, 62%; range, 0.083 to  $0.37 \mu\text{g L}^{-1}$ ; outliers

TABLE 2. Concentrations of Cd, Pb, and Cu in waters collected at the Sibylline Mountains National Park, central Italy

Sample, date	Metal concn <sup>a</sup>		
	Cd (ng L <sup>-1</sup> )	Pb (ng L <sup>-1</sup> )	Cu (µg L <sup>-1</sup> )
<b>Mount Bove north</b>			
Springs			
Panico, July 2008	1.7	16	0.10
Val Ruscio, July 2009	1.1	20	0.31
Val Ruscio, June 2011	1.2	20	0.31
L'Acqua Fiecciola, July 2009	1.6	18	0.14
Bottled mineral water			
Roana, Nov. 2008	0.8	16	0.10
Aqueduct waters			
Frontignano public fountain, Nov. 2004	1.1	15	0.13
Frontignano public fountain, July 2005	1.2	17	0.14
Frontignano public fountain, July 2008	1.3	16	0.15
Ussita public fountain, July 2008	1.5	25	0.37
Ussita public fountain, June 2011	1.4	21	0.36
Sorbo rural house, July 2009	4.4 <sup>b</sup>	104 <sup>b</sup>	0.65 <sup>b</sup>
Sorbo rural house, June 2011	2.0 <sup>b</sup>	101 <sup>b</sup>	0.61 <sup>b</sup>
Mean ± SD (RSD%)	1.2 ± 0.3 (25)	18 ± 3 (17)	0.21 ± 0.11 (52)
<b>Mount Bove south</b>			
Springs			
Fonte del Lupo, July 2005	1.0	6	0.10
Fonte del Lupo, July 2008	1.2	6	0.13
Fonte del Lupo, June 2011	1.1	7	0.10
Val di Bove, June 2011	1.2	8	0.12
Aqueduct water			
Rifugio Saliere mountain hut, Mar. 2011	1.2	10	0.30 <sup>b</sup>
Mean ± SD (RSD%)	1.1 ± 0.1 (9)	7 ± 2 (22)	0.11 ± 0.02 (18)
<b>Springs of the River Nera</b>			
Springs			
Uccelletto, July 2008	1.1	7	0.11
Acero, July 2008	1.4	18	0.10
Nera, July 2005	1.2	10	0.083
Nera, July 2008	2.1	9	0.10
Bottled mineral water			
Nerea, July 2008	2.4	11	0.11
Aqueduct water			
Vallinfante public fountain, July 2008	0.7	13	0.10
Mean ± SD (RSD%)	1.5 ± 0.6 (40)	11 ± 4 (36)	0.10 ± 0.01 (10)
Overall mean ± SD (RSD%)	1.3 ± 0.4 (31)	14 ± 6 (43)	0.16 ± 0.10 (62)

<sup>a</sup> Each value represents the mean of at least three measurements for different aliquots of the same sample. Measurement variabilities expressed as pooled standard deviations are 19% for Cd, 15% for Pb, and 8% for Cu.

<sup>b</sup> Outliers not considered for summary statistics.

excluded) (Table 2). All concentrations were well below the legal limit (1 µg L<sup>-1</sup>) stipulated by national (30–34) and international (15–17, 19, 54, 55, 57) institutions (Table 1).

Differences in Cu concentrations were found among the three areas studied. Concentrations were similar for the River Nera springs (0.10 ± 0.01 µg L<sup>-1</sup>) and Mount Bove south (0.11 ± 0.02 µg L<sup>-1</sup>), but the concentration was double at Mount Bove north (0.21 ± 0.11 µg L<sup>-1</sup>). In this area, differences between springs were evident, with a substantial

split in two zones. One zone included the Panico and Acqua Fiecciola springs, with an overall mean Cu concentration of 0.12 ± 0.02 µg L<sup>-1</sup>, and the other included the Val Ruscio spring, with a threefold higher overall mean of 0.31 µg L<sup>-1</sup>. One possible explanation for these differences may be the different materials used in the distribution pipeline. This difference is clearly evident in a comparison of spring and aqueduct waters. In the rural house in Sorbo, Cu concentration was 0.65 µg L<sup>-1</sup>, six times higher than that

TABLE 3. Comparison of our results with previously published data for metal concentrations in tap and mineral waters

Type of sample	Statistic	Cd (ng L <sup>-1</sup> )	Pb (ng L <sup>-1</sup> )	Cu (μg L <sup>-1</sup> )	Reference
Bottled mineral water					
Roana brand (central Italy)	Mean ± RSD <sub>pooled</sub>	0.8% ± 19%	16% ± 15%	0.10% ± 8%	This study
Nerea brand (central Italy)	Mean ± RSD <sub>pooled</sub>	2.4% ± 19%	11% ± 15%	0.11% ± 8%	This study
Italian (186 brands)	1°–3° quartile	2–7	3–13	0.12–0.30	10
European (1,785 brands)	1°–3° quartile (median)	<3–8 (3)	<10–720 (93)	0.13–0.72 (0.25)	38
Alabama (25 brands)	Range	<0.2 × 10 <sup>3</sup> –5 × 10 <sup>3</sup>	<LOD–3 × 10 <sup>3</sup>	<0.2–2	24
Egyptian (5 brands)	Mean ± RSD%	10 ± 1.39	80 ± 2.45	10.5 ± 1.39	40
Tap water					
Mount Bove north (central Italy) <sup>a</sup>	Mean ± RSD <sub>pooled</sub>	1.3% ± 19%	19% ± 15%	0.23% ± 8%	This study
Mount Bove South (central Italy)	Mean ± RSD <sub>pooled</sub>	1.2% ± 19%	10% ± 15%	0.30% ± 8%	This study
Springs of the River Nera (central Italy)	Mean ± RSD <sub>pooled</sub>	0.7% ± 19%	13% ± 15%	0.10% ± 8%	This study
Italian tap water	1°–3° quartile	1.3–14.6	26–288	0.91–6.37	13
European tap water	1°–3° quartile (median)	4–17 (8)	41–280 (120)	2.2–17 (5.7)	38
Egypt tap water (Giza City)	Mean ± RSD%	30 ± 2.12	210 ± 1.71	4.46 ± 0.51	40
Egypt tap water (Cairo City)	Mean ± RSD%	50 ± 21.9	140 ± 23.5	4.72 ± 23.7	40
Pakistan tap water (Charsadda District; tube well)	Mean ± SD	2.3 × 10 <sup>3</sup> ± 0.19 × 10 <sup>3</sup>	9.9 × 10 <sup>3</sup> ± 8.2 × 10 <sup>3</sup>	15.5 ± 1.1	28
Spring water					
Mount Bove north (central Italy)	Mean ± SD	1.4 ± 0.3	18 ± 2	0.22 ± 0.11	This study
Mount Bove South (central Italy)	Mean ± SD	1.1 ± 0.1	7 ± 1	0.11 ± 0.02	This study
Springs of the River Nera (central Italy)	Mean ± SD	1.4 ± 0.5	11 ± 5	0.10 ± 0.01	This study
Springs of Mount Reventino (southern Italy)	1°–3° quartile	<20	<60–100	<0.1–0.53	4
Ishikawa Prefecture (Japan)	Mean ± SD	18 ± 8			5

<sup>a</sup> Outliers (data from house in Sorbo) were excluded from the analysis.

for the Panico spring and two times higher than that for the Val Ruscio spring. Corrosion and interaction with domestic water distribution systems can affect the concentrations of Cu in tap water (13, 20). No other aqueduct samples in this study had similar Cu concentrations, supporting the distribution system hypothesis and confirming that the last mile of the water system is the most critical part for water contamination. The Roana and Nerea bottled mineral waters had the same Cu concentrations as did their springs (Panico and Uccelletto, respectively), indicating that the bottling procedures were free of Cu contamination.

The Cu concentration of Sibylline mineral water is in agreement with values reported previously for Italian and European mineral waters (10, 37) and lower than those reported in other countries (24, 39). The Cu concentrations measured for tap water for all areas in the present study were 5- to 90-fold below all reported Italian and European Cu concentrations (Table 3) (13, 27, 37, 39). The Cu concentrations in spring waters were basically similar to the few reported values, indicating the good quality of these spring waters.

All waters of the Sibylline Mountains National Park analyzed in this study were of good quality and could be classified (according to Italian law) as oligomineral waters of medium to discrete hardness that are suitable for low-sodium diets and for infant consumption.

The Cd, Pb, and Cu concentrations were well below Italian and European legal limits and standards for drinking waters from other authorities: the Environmental Protection Agency (EPA), World Health Organization (WHO), and Food and Agriculture Organization of the United Nations (FAO). The bottling process, with few exceptions, did not influence the concentrations of heavy metals; no significant differences in concentrations were found between the springs and the corresponding bottled mineral waters.

Differences in metal concentrations were found between springs and aqueduct waters in a rural house. The increased concentrations of all metals were probably related to the old pipes forming the distribution line. This finding suggests that it is necessary to pay great attention to the material used in the water systems of older private houses.

This study revealed the lack of pollution in the waters of the Sibylline Mountains National Park, the most important

water reserve of Marche region of Italy, and paves the way for similar studies, including future monitoring of these characteristics, in this region recently hit by a catastrophic earthquake. This area will be a key supplier of drinking water for other areas of central Italy in the future.

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### SUPPLEMENTAL MATERIAL

Supplemental material associated with this article can be found online at: <https://doi.org/10.4315/0362-028X.JFP-17-310.s1>.

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