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Evaluation of Selected Metal Elements in Commercial Drinking Water and Tap Water in Peninsular Malaysia

(Penilaian terhadap Elemen Logam Terpilih dalam Air Minuman Komersil dan Air Paip di Semenanjung Malaysia)

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ABSTRACT

The present study was carried out to determine the concentrations of selected metal elements (lead, copper, manganese, zinc and iron) in 51 samples of commercial drinking water and tap water available in Malaysia. The results indicated that low metal elements were found in the studied water samples. Lead, manganese, zinc and iron were not detected in some of the studied samples, except copper. The concentrations of the metal elements in the studied samples were well below the maximum permitted concentrations as recommended. Therefore these drinking water are safe for consumption and do not pose adverse effect to the health of consumers due to metal toxicity.

Keywords: Metal elements, Drinking water, Mineral water, Tap water, Health

ABSTRAK

Kajian ini dijalankan untuk menentukan kandungan unsur logam yang terpilih (plumbum, gangsa, mangan, zink dan besi) dalam 51 sampel air minuman komersil dan air paip yang sedia ada di Malaysia. Keputusan kajian mendapati bahawa kandungan unsur logam adalah rendah dalam sampel air yang dikaji. Plumbum, mangan, zink dan besi tidak terdapat dalam sebilangan kecil sampel yang dikaji, kecuali kuprum. Kandungan unsur-unsur logam dalam sampel yang dikaji adalah di bawah kandungan minima yang dibenarkan seperti yang disarankan. Oleh itu, air minuman yang dikaji adalah selamat untuk diminum dan tidak mendatangkan kesan buruk kepada kesihatan pengguna disebabkan oleh keracunan logam.

Kata kunci: Unsur logam, Air minuman, Air mineral, Air paip, Kesihatan

INTRODUCTION

Water is the most vital substance for life and contains essential minerals. Water quality is crucial to be maintained to ensure safety and causing no harm for drinking. Safe drinking water should be free from harmful levels of impurities such as bacteria, viruses, heavy metals and toxic organic substances (Gadgil 1998). The water must also be free from unpleasant tastes and odor. Water contamination is known to occur due to geological phenomena such as ore formations, human activities, agricultural and industrial applications, atmospheric fall out, soil erosion and deforestation (Al Fraij et al. 1999; Sillanpää et al. 2004).

Metal elements are harmful to humans due to their long biological half lives and prone to accumulate in the body (Ikem et al. 2002; Pyatt et al. 2005; Watt et al. 2000). The typical heavy metal elements found in water sources polluted by industrial wastes are nickel, cadmium, mercury and lead (Gadgil 1998). Other metal elements (copper, manganese, zinc and iron) are also found in industrial waste water. Most of these metals are freely dissolved in water and extremely toxic (Awofolu et al. 2005). It was reported that low concentrations of heavy metals in blood has damaging effects such as reduced growth and development in amphibians (Blaustein et al. 2003). Therefore, the concentrations of metal elements in commercial drinking water and tap water should be determined to enlighten people on the quality of available commercial drinking water and tap water.

In Malaysia, all commercial drinking water should comply with the Food Regulations 1985, read together with the Food Act 1983. In these regulations, commercial drinking water is classified under Regulation 360A for Natural Mineral Water and Regulation 360B for Packaged Drinking Water (Legal Research Board 2008). In accordance with regulation 360A(1) of the Food Regulations 1985, natural mineral water is the ground water obtained for human consumption from various sources. Regulation 360B(1) of the Food Regulations 1985 defined packaged drinking water (which includes reverse osmosis water) as potable or treated potable water that is filled in bottles or other packages and for the purpose of human consumption. In addition, the WHO (2006) guideline is applied to determine the safety and quality of commercial drinking water. On the other hand, the quality of tap water is very much dependent on hygiene and quality management of the water during its treatment and distribution (Jain & Singh 2003).

The present study was conducted to determine the concentrations of selected metal elements (lead, copper, manganese, zinc and iron) in drinking water in Malaysia as toxicities related to these elements can result in major health problems to individuals. The concentrations of metal elements determined were compared to the existing international and national standards.

MATERIALS AND METHODS

SAMPLES AND SAMPLING

A total of 51 samples comprising reverse osmosis water (ROW) (n = 14), mineral water (MW) (n = 13) and tap water (TW) (n = 24) were obtained from various towns and cities in Peninsular Malaysia. TW samples were collected from two different locations in 12 states of West Malaysia each in May and June, 2009. The 24 TW samples were from Perlis (TW 1 & TW 2), Kedah (TW 3 & TW 4), Pulau Pinang (TW 5 & TW 6), Perak (TW 7 & TW 8), Selangor (TW 9 & TW 10), Wilayah Persekutuan (TW 11 & TW 12), Negeri Sembilan (TW 13 & TW 14), Melaka (TW 15 & TW 16), Johor (TW 17 & TW 18), Pahang (TW 19 & TW 20), Terengganu (TW 21 & TW 22), and Kelantan (TW 23 & TW 24). As indicated in Figure 1, the two different locations from each state for water sampling were selected based on random sampling method. Two replicates of sample from each source were collected in plastic containers that had been treated with 0.01 M HNO_3 and rinsed with ultra-pure water. One litre samples were collected from the tap after the water was left to run for at least 5 min before collection. The samples were kept in sealed plastic bottles, transported to laboratory and stored at 4-6 C.

The samples of commercial drinking water (27 brands) were purchased from local supermarkets and hypermarkets in the Klang Valley, Malaysia in May–June, 2009. Stratified sampling was applied during sample collection, where the bottled drinking waters were randomly selected from the shelves of six identified supermarkets and hypermarkets. The drinking water samples were stratified according to supermarkets and hypermarkets that are selling drinking water, where "different brands of drinking water are not expected to show significant variation" (Greenfield & Southgate 2003). The samples comprised 13 brands of MW and 14 brands of ROW. All of the commercial drinking water samples were packed in sealed plastic (PET) bottles (500 mL). The samples were stored at 4 C before analysis. All samples were analyzed within 14 days from the day of collection and no preservatives were added.

SAMPLE AND STANDARD PREPARATION

TW samples were filtered through a 0.45 µm membrane filter before analysis. The pH of the filtrate was set to 2 ± 0.2 with 1 M HNO_3 using a Toledo 320 pH meter (Mettler-Toledo Inc. Columbus, OH). Standard solutions of metals (1000 mg/L) were purchased from Merck (Darmstadt, Germany). Stock solutions of metal standards were prepared by dilution with ultra-pure water from Milli-pore water system (Millipore, Bedford, MA, USA). Working solutions of the standards were prepared daily by diluting the stock solutions with ultra-pure water. Five different concentrations of standard solutions (0.1–5 mg/L) were prepared to determine the standard calibration. To ensure the water samples were not contaminated during sample preparation, all plastic containers and glass wares were treated with 0.01 M HNO_3 and rinsed with ultra-pure water.

METAL ELEMENT ANALYSIS

Metal element concentrations in 100 mL of the drinking water samples were determined using a novAA 400 flame atomic absorption spectrophotometry (AAS) (AnalytikJena, Jena, Germany) and a GBC 908AA graphite furnace AAS (GBC, Victoria, Australia). Flame AAS was used to determine the concentrations of Fe, Cu and Zn in drinking water samples, while graphite furnace AAS was applied to determine the concentration of Pb in the studied samples. The concentration of Mn in the water samples were analyzed using inductively coupled plasma-optical emission spectrophotometry (ICP–OES) system equipped with auto sampler, electrothermal vaporization, laser ablation, ultrasonic nebulizer and hydrite generation system (Hewlwtt-Packard Co. Wilmington, DE).

The analysis was carried out based on EPA Method 200.8 (EPA 1995) and method described by Chen et al. (1995). For AAS analysis, a range of wavelength (213.9–766.5 nm) was applied in metal element analysis. The accuracy of the analysis was performed using Certified Reference Materials (CRM-616 and CRM-617) from Commission of European Communities. In the drinking water samples, the lowest concentration of metal elements that can be quantified using AAS were 0.10 µg/L for Fe, Cu and Zn, while 0.09 for Pb µg/L. The quantification limit for Mn using ICP–OES was 1.0 µg/L. The linearity of the standard calibrations for Fe, Cu, Zn, Pb and Mn were determined by the R^2 of higher than 0.99. All data were statistically analysed using Minitab Version 15. The significant values of the data obtained were statistically analysed using one-way analysis of variance (ANOVA) with p value set at 0.05. Tukey's analysis was applied for post-hoc comparison.

RESULTS AND DISCUSSION

METAL ELEMENT CONCENTRATIONS

The concentrations of metal elements in the drinking water samples obtained in the study varied from "Not Detectable" to 330.00 µg/L (Fe), 358.2 µg/L (Zn), 3.92 µg/L (Pb) and 91.00 µg/L (Mn), except for Cu (0.2–26.3 µg/L). Table 1 shows the metal element concentrations in the drinking water samples. The results show that TW samples contained the highest concentrations of Fe, Zn, Pb and Mn, while MW samples were found to have the highest Cu concentration compared to the other samples studied. The results also showed that metal elements were detected in all types of drinking water. All studied drinking water samples have Fe and Mn concentrations within the range determined using CRM 616 and CRM 617.

Malaysia. The non-significant differences for metal elements found among the 12 states of Peninsular Malaysia were due to the large variation of metal element concentrations taken from two different locations in each state. The results from post-hoc comparison also showed that the concentrations of metal elements between and among some of the ROW and MW samples were not significantly different.

The ROW samples were found to contain relatively lower concentrations of metal elements than MW except for Fe and Pb. However, the concentrations of metal elements found in the samples were below the maximum permitted concentrations set by the Legal Research Board (2008) and WHO (2006) (Table 2). Pb was not detected in most of the TW samples, except TW 9, TW 23 and TW 24. The results are in agreement with Güler and Alpaslan (2009) that some of the commercial mineral water sold in the

TABLE 1. Metal elements in commercial reverse osmosis water, mineral water and tap water

Type of water		Fe	Cu	Zn	Pb	Mn
Reverse osmosis water	Mean (µg/L)	34.51	2.99	1.31	1.28	5.14
	SD	7.34	2.50	0.79	1.33	8.81
	Max	43.30	7.30	2.70	3.92	31.00
	Min	17.20	0.20	0	0	0
	N	14	14	14	14	14
	Variation (%)	21.3	83.6	60.3	103.9	171.4
Mineral water	Mean (µg/L)	11.62	12.77	4.79	0.26	31.54
	SD	16.88	3.92	6.75	0.37	21.44
	Max	60.50	16.90	24.30	1.25	68.00
	Min	0	1.50	0.40	0	0
	N	13	13	13	13	13
	Variation (%)	145.3	30.7	140.9	142.3	68.0
Tap water	Mean (µg/L)	70.00	8.59	34.70	0.32	25.39
	SD	61.92	6.09	72.83	0.94	22.43
	Max	330.00	26.30	358.20	3.80	91.00
	Min	23.20	0.90	2.1	0	0
	N	24	24	24	24	24
	Variation (%)	88.46	70.9	209.9	293.8	88.3
CRM 616	Mean (µg/L)	50	–	–	–	18
CRM 617	Mean (µg/L)	206	–	–	–	50

SD = standard deviation; Max = maximum; Min = minimum; n = sample size; percentage of variation is calculated based on the following formula: (SD/Mean) × 100; variation (%) is also known as RSD (%) (AOAC 2002).

The concentrations of metal elements in different brands of commercial drinking water (ROW and MW) and in various sources of TW are shown in Tables 2 and 3. There were significant differences among the metal elements for ROW and MW samples studied ($p < 0.001$). No significant different was found for metal elements in TW samples among the 12 states studied; except for Pb with $p < 0.05$. The post-hoc comparison showed that the Pb concentrations in TW 23 and TW 24 obtained from Kelantan were significantly different from other states in Peninsular

Turkish market has non-detectable amount of Fe, Zn and Mn.

In this study, the concentrations of metal elements in the drinking water samples were much lower than the values reported by Rosborg et al. (2005), except for Fe. Although various adverse effects have been reported related to the intake of metal elements through oral means, some common adverse effects of exposure to metal element are cancer, adverse reproductive outcomes, cardiovascular and neurological diseases (Watt et al. 2000).

TABLE 2. Metal element concentrations (µg/L) in commercial drinking water sample

Item	Fe**	Cu**	Zn**	Pb**	Mn**
ROW 1	17.20	1.70	ND	3.80	ND
ROW 2	23.40	0.20	0.60	1.50	ND
ROW 3	41.30	1.00	0.80	1.81	ND
ROW 4	42.10	0.60	2.70	1.57	ND
ROW 5	35.50	0.40	1.80	2.34	1.00
ROW 6	37.90	1.20	1.40	0.66	ND
ROW 7	28.70	1.00	1.80	0.51	ND
ROW 8	43.30	2.80	2.00	ND	2.00
ROW 9	37.00	3.60	1.40	ND	31.00
ROW 10	36.70	4.10	1.50	0.28	15.00
ROW 11	38.40	5.30	1.30	3.92	11.01
ROW 12	30.20	5.70	0.30	ND	ND
ROW 13	36.60	6.90	0.50	1.34	12.01
ROW 14	34.80	7.30	2.30	0.24	ND
MW 1	16.70	12.30	4.10	ND	ND
MW 2	17.40	10.10	3.30	0.09	19.02
MW 3	19.20	11.60	1.40	0.33	35.01
MW 4	17.30	12.60	24.30	ND	39.01
MW 5	15.90	12.20	12.70	0.69	30.90
MW 6	60.50	13.00	3.40	0.44	67.00
MW 7	3.00	14.80	2.20	ND	4.00
MW 8	0.10	14.30	1.20	ND	6.00
MW 9	ND	15.10	0.80	1.25	39.02
MW 10	ND	16.40	1.20	0.28	68.00
MW 11	ND	1.50	0.70	0.34	30.00
MW 12	ND	16.90	0.40	ND	46.00
MW 13	ND	15.20	6.60	ND	26.02
WHO Guidelines 2006	–	2000	–	10	400
Malaysian Food Regulations 1985 (360A)	–	1000	5000	50	2000
Malaysian Food Regulations 1985 (360B)	300	1000	5000	50	100

ND = not detected; ROW = reverse osmosis water; MW = mineral water; TW = tap water; ** Significant difference at $p < 0.001$.

TABLE 3. Metal element concentrations (µg/L) in tap water sample

Item	Fe	Cu	Zn	Pb*	Mn
TW 1	56.40	6.20	19.60	ND	11.10
TW 2	31.10	9.30	25.10	ND	20.00
TW 3	51.60	4.00	17.30	ND	4.01
TW 4	59.50	2.00	3.60	ND	29.01
TW 5	29.60	2.80	5.40	ND	34.01
TW 6	28.00	1.80	2.10	ND	23.02
TW 7	63.50	1.90	11.10	ND	ND
TW 8	85.30	0.90	12.20	ND	14.01
TW 9	33.30	5.60	13.10	2.49	4.01
TW 10	87.40	7.40	358.20	ND	40.01
TW 11	85.50	7.60	56.70	ND	12.03
TW 12	31.80	13.00	17.80	ND	6.00
TW 13	29.80	5.60	21.60	ND	27.00
TW 14	32.20	4.50	4.50	ND	91.00
TW 15	33.80	5.30	4.80	ND	32.01
TW 16	68.90	19.90	75.30	ND	55.00
TW 17	23.20	8.50	18.70	ND	30.00
TW 18	58.20	10.50	96.70	ND	46.01
TW 19	31.00	10.90	7.70	ND	47.01
TW 20	47.60	26.30	17.20	ND	ND
TW 21	61.20	13.30	4.30	ND	ND
TW 22	31.20	12.20	4.10	ND	33.00
TW 23	330.00	12.60	30.80	3.80	ND
TW 24	44.00	14.00	4.80	1.50	51.00
WHO Guidelines 2006	–	2000	–	10	400
Malaysian Food Regulations 1985 (360A)	–	1000	5000	50	2000
Malaysian Food Regulations 1985 (360B)	300	1000	5000	50	100

ND = not detected; ROW = reverse osmosis water; MW = mineral water; TW = tap water; * Significant difference at $p < 0.05$; Fe, Cu, Zn and Mn for the TW in were not significantly different between and among the 12 states studied.

VARIATIONS OF METAL ELEMENT CONCENTRATIONS AMONG DIFFERENT SOURCES AND LOCATIONS

The concentrations of metal elements in ROW, MW and TW samples varied between samples and between locations. The samples were collected from various locations in Peninsular Malaysia (Figure 1). Less than 50% variations were found for Fe and Cu concentrations among ROW and MW samples collected from various water sources. More than 100% variations were found for Pb and Mn concentrations in ROW samples, for Fe, Zn and Pb concentrations among MW samples, Zn and Pb concentrations among TW samples. About 20% variation was found for Fe concentrations among ROW samples. However, larger variations were found for Zn and Pb among TW samples compared to ROW and MW samples. Therefore, the geographical location of the water sources could be the factor that affects the metal content, especially heavy metal elements.

The results indicate that although similar types of water were analyzed, the concentrations of metal elements varied between and among ROW, MW and TW samples. TW samples showed the greatest variation in metal element concentrations due to the water obtained from various sources with different geographical locations (Figure 1). Although ROW and MW were purchased from different supermarkets and hypermarkets in different places, most of the ROW and MW originated from a few similar sources. Previously, it has been reported that MW obtained from ground water, originated from mining and industrial zones, are contaminated by heavy metals (Chatterjee et al. 1993; Pye & Patrick 1983). Similarly, in some samples of this study also showed higher lead concentration, especially in MW 9, ROW 1 and ROW 5.

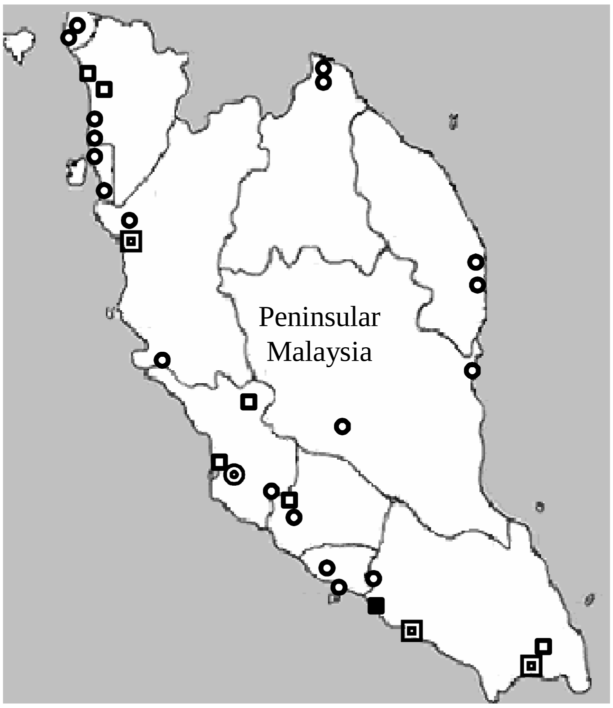

FIGURE 1. Sources and locations of drinking water samples

The square and circle spots are the drinking water collected from different locations:
□ = Tap water, ▣ = Mineral water, ○ = Drinking water, ■ = Tap water + Mineral water, ▣ = Tap water + Drinking water, ◉ = Mineral water + Drinking water.
The map of Peninsular Malaysia is adapted from PETRACK (2008).

Water treatment and filtration processes before packaging of drinking water also might affect the metal compositions in ROW and MW (Alpatova et al. 2004). Besides, the TW from different locations was treated using different approaches before being supplied to consumers; while ROW and MW were treated using standard treatment and filtration procedures. During the water filtration process for commercial drinking water, most of the metal elements are removed but small amount of metal elements can pass through the filter system. Therefore, small variations of metal element concentrations among ROW and MW samples were found compared to TW samples.

COMPARISON OF METAL ELEMENT CONCENTRATIONS WITH MAXIMUM PERMITTED VALUES

In TW samples, Fe concentrations from various places in Malaysia were higher than the concentrations found in commercial drinking water samples. TW 23 had the highest concentration of Fe (330.00 µg/L), followed by TW 10 (87.40 µg/L), TW 11 (85.50 µg/L), etc. Compared to the study by Rosborg et al. (2005), the mean value of Fe (23.07 µg/L) in commercial drinking water (ROW and MW) in this study was higher than the mean value of Fe (2.0 µg/L) found in bottled water in the Swedish market. Although many of the main underground water pipes in Malaysia had been replaced by MDPE, the water pipes and some of household water pipes near outlets are made from iron. Therefore, the high concentration of iron in some studied TW samples could be due to the presence of rust in the corroded water piping system as supported by Colter and Mahler (2006) that "ferric iron deposited within the corroded pipes can break free and generate rusty tap water."

In the WHO Guidelines for Drinking-Water Quality (WHO 2006), no specific maximum permitted concentration has been set for Fe in drinking water. Previously, WHO (1993) guideline had set a maximum permitted concentration below 2 mg/L, as did not pose any hazard to health. Generally, the concentration of Fe in the samples studied does not exceed 0.12 mg/L, which is the threshold value for Fe in water containing dissolved solids (Saleh et al. 2001). On the other hand, the Fe concentration in commercial drinking water samples were relatively lower and below the permitted concentration (0.3 mg/L) stated in the Malaysian Food Regulations 1985 (Legal Research Board 2008), except for TW 23 sample (0.33 mg/L) (Table 2). Nevertheless, iron toxicity seldom occurs through consumption of TW or other commercial drinking water.

In the samples studied, the mean values for Cu were relatively higher in both MW (12.77 µg/L) and TW (8.59 µg/L) compared to ROW (2.99 µg/L). Although Cu was present in most of the samples, its concentration was very much lower than the recommended maximum permitted concentration (WHO 2006 and Legal Research Board 2008), at 2 mg/L and 1 mg/L respectively (Table 2). However, the Cu concentrations in commercial drinking water samples were comparable to the bottled drinking water from Egypt (Saleh et al. 2001). Despite Cu being an essential mineral, the elevated concentrations of Cu may result in vomiting, nausea, diarrhea, liver and kidney damages, and gastrointestinal problems (Pyatt et al. 2005; Sandstead 1995). Cu toxicity from dietary sources is extremely rare, but can result from drinking contaminated water or consuming acidic foods or beverages stored in copper containers (Olivares et al. 2000).

The concentrations of Zn in commercial drinking water samples were very much lower than the concentrations detected in TW samples. In TW samples, the highest Zn concentration was found in TW 10 (358.20 µg/L), followed by TW 18 (96.70 µg/L), TW 16 (75.30 µg/L), etc. No specific maximum permitted concentration for Zn has been set by the WHO Guidelines for Drinking-Water Quality (WHO 2006), but the permitted concentration in the Malaysian Food Regulations 1985 (Legal Research Board 2008) is 5 mg/L (Table 2). As all drinking water samples had Zn concentrations below the maximum permitted concentration, the result indicates that the samples studied are safe.

In this study, Pb was found in relatively higher concentrations in ROW compared to the other samples studied, but the concentrations were still below the maximum permitted concentration recommended by WHO (2006) and Malaysian Food Regulations 1985 (Legal Research Board 2008), where the maximum permitted concentrations are 10 µg/L and 50 µg/L, respectively (Table 2). Although Pb was not detected in most of the drinking water samples, 11 brands of commercial drinking water and three sources of TW samples were found to contain low concentrations of Pb. As Pb is rarely found in ground water, the detectable concentrations of Pb in drinking water samples is suspected to be contamination from corrosion of plumbing materials that contain lead (Schock & Neff 1988).

The results indicate that ROW samples contained Mn ranging from 0–31.00 µg/L, while 0–68.00 µg/L of Mn was found in MW samples, and 0–91.00 µg/L in TW samples. The highest concentration of Mn was found in TW sample from TW 14 (91.00 µg/L), followed by TW 16 (55.00 µg/L), TW 24 (51.00 µg/L), etc. Among the MW samples, MW 6 and MW 10 have high Mn concentrations, i.e. 67.00 and 68.00 µg/L, respectively. Even though the samples were found to contain high levels of Mn, the concentrations of were still below the maximum permitted concentration recommended by WHO guideline (WHO 2006) and Malaysian Food Regulations 1985 (Legal Research Board 2008) (Table 2).

CONCLUSION

Generally, all of the water samples studied contains metal elements such as Fe, Cu, Zn, Pb and Mn. However, their concentrations were much lower than the maximum permitted concentrations recommended by the WHO Guidelines for Drinking-Water Quality 2006, Regulation 360A (Natural Mineral Water) and Regulation 360B (Packaged Drinking Water) of the Malaysian Food Regulations 1985. Therefore, commercial drinking water (reverse osmosis and mineral water) and tap water in Malaysia are safe for human consumption.

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