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Assessment of the Altitudinal Atmospheric Metal(loid) Deposition in a Mountainous City by Mosses

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Abstract Samples of moss (*Haplocladium microphyllum*) were collected at different elevations on a mountain and four representative sites in Guiyang City, and the concentrations of metal(loid)s were determined by ICP-MS. The altitudinal deposition of soil-originated metals differed from that of anthropogenic metal(loid)s. The concentrations of soil-related elements decreased with elevation, indicating that these elements tend to deposit at lower elevations and their impact on the higher elevations is less. The concentrations of anthropogenic elements varied only slightly with elevation, indicating that the atmospheric deposition of these elements did not vary largely with elevation. The results of this study showed that the mosses at different locations may serve to indicate a vertical gradient of atmospheric metal(loid) deposition.

Keywords Metals · Metalloids · Deposition · Moss · Vertical gradient

Atmospheric metal(loid) deposition has become a major concern in urban environments. Emissions from natural and anthropogenic sources contributed increasing amounts of metal(loid)s into the atmosphere over the last decade, resulting in poor air quality and adverse health effects (Cheng et al. 2012). Thus, the monitoring of airborne metals has become an essential part of environmental planning and control programs in many parts of the world (Sella et al. 2001; Cayir et al. 2007). Due to the gravity of size-related particles, the distribution of both particles and contaminants can vary largely with elevation (Naimi and Ayoubi 2013). However, moss biomonitoring along the vertical gradient of contaminants has only been carried out in some remote areas. The metal concentrations of mosses have been observed to vary significantly with increasing elevation in the Alps (Zechmeister 1995; Gerdol et al. 2002; Gerdol and Bragazza 2006), the Tatra Mountains in Slovakia (Soltes 1998), and the Nanling Mountains in China (Lee et al. 2005).

In cities, the exposure of inhabitants in high buildings to air pollution may differ owing to the gradient of atmospheric contaminants, making it significant to understand the vertical dispersion of contaminants in urban environments. Moss contaminant analysis at different altitudes is able to provide valuable information to characterize contaminant deposition and to ultimately develop appropriate air pollution control strategies. However, this type of research has rarely been reported.

Guiyang City is the capital of Guizhou province in southwest China. This city is located in a mountainous area, and the genus *Haplocladium microphyllum* occurs widely in the urban area even on mountains, making it possible to evaluate the vertical gradient of metal(loid) deposition using moss biomonitoring. *H. microphyllum* has been shown to be a good biomonitor of air quality (Cao et al. 2008). The objective of this study was to utilize the epigaeic moss species *H. microphyllum* from different elevations of a mountain to assess the altitudinal metal (loid) deposition in an urban environment.

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Materials and Methods

Guiyang City (26°34′N, 106°43′E) is situated in a karst basin (elevation 1050 m) and is surrounded by hills and mountains. The city has an urban area of 220 km^2 and a population of ca. 4 million people. Guiyang has a subtropical monsoon climate with an annual average temperature of 15.3°C, an annual average rainfall of 1174 mm, and an average relative humidity of 86 %. Prevailing winds are northeastern, and the secondary wind direction is from the south (Fig. 1). Annual wind speed is 2.2 m/s. Shuikousi Mountain (SKS Mt.) is located in the eastern part of Guiyang. This mountain has a steep average slope of 50°, and the peak elevation is 222 m from the ground. Its vegetation is characterized by locust trees and grasses. There is a road along the base of the mountain that has dense traffic. Two important industrial factories in the area of Guiyang, Iron & Steel Works (ISW) and Aluminum Plant (AP), are 3 km south and 20 km northwest of SKS Mt., respectively. As with many other cities across China, many large construction projects are being carried out to lessen the housing shortage caused by rapid urbanization, so the air is increasingly influenced by the construction-derived dust. In addition, the quantity of vehicles was 750,000 in 2011 and is increasing by ca. 150 vehicles per day, so road traffic is becoming an important source of contaminants in the atmosphere. A construction site (CS) in suburban Guiyang and a road with busy traffic (Bihai road, BR) were selected for evaluation of soil dust and traffic-related influences, respectively.

Based on its high presence, quick growth rates, and high pollution endurance, *H. microphyllum* was selected for monitoring metal(loid) deposition. Mosses were collected at SKS Mt. and four representative metal(loid) source sites (ISW, AP, CS, and BR) (Fig. 1). Mosses within a 1 m^2 area were collected and combined into one representative sample to avoid microclimate influence. Samples were collected from natural moss layers (4–5 cm thickness) located above ground level (to avoid surface water splashes), and sites potentially disturbed by domestic animals or pets were avoided. Four to five moss samples were collected at each representative source site. At SKS Mt., a total of 29 moss samples were collected along a steep slope from 1098 to 1256 m a.s.l., which corresponds to 64–222 m above the base of the mountain.

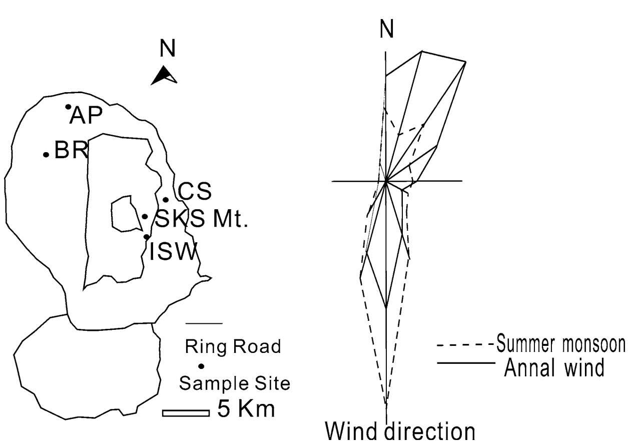

Fig. 1 Map showing the sampling sites and the wind rose plot

The preparation of moss samples in laboratory was performed following the modified method of Lee et al. (2005). Dead material and litter were removed from the samples. The green part of moss tissue was separated and soil particles adhered on the surfaces were removed. The samples were dried in an oven at 50°C for 3 days. For the digestion of samples, 50 mg of mosses was weighed and placed into a Teflon crucible (ID: 30 mm, height: 55 mm). Then, 1 mL of sub-boiling distilled nitric acid (HNO_3) and 0.2 mL of sub-boiling distilled hydrofluoric acid (HF) were added. The crucible was sealed within a stainless steel bomb and heated at 140°C (internal pressure: ca. 3.6 bar) for 12 h. After cooling, the digestion bomb was opened and heated at 80–90°C on an electrothermal plate to remove the excess HF. Then the residue was re-dissolved in a 50 mL volumetric flask with 1 % HNO_3, and stored in the dark at 4°C.

Metal(loid) determination was conducted by a quadrupole ICP-MS (Platform ICP, Micromass Inc., Manchester, UK). The plasma power of the instrument was 1350 w, and the rates of cool Ar gas flow, auxiliary Ar gas flow, and nebulizer Ar gas flow were 13.5, 1.2, and 0.7 L/min, respectively. The sample flow rate was 1 mL/min. Method blanks, certified reference poplar leaves (GBW07604, China National Research Center for CRM's, Beijing, CN) were digested and analyzed at least in triplicate. Accu-Trace ICP-MS standard solutions of 10 μg/mL for each element (AccuStandard Inc., New Haven, CT, USA) were diluted in a matrix of 1 % HNO_3 for the external calibration of analysis. Detection limits of the instrument and recoveries for the metal(loid)s in certified reference material are presented in Table 1.

SPSS 16.0 software (SPSS Inc., New York, USA) was used for statistical analysis. Regarding the metal(loid) concentrations of mosses from the four representative sites, the Shapiro–Wilk test was used to assess the normality of the distributions of metal(loid) concentrations in mosses at each site (the data were log-transformed when necessary) and Levene's test was used to assess the homogeneity of variances. The mean values of metal(loid) concentrations in mosses from the four representative sites were checked with a one-way ANOVA followed by Tukey-HSD test ($p < 0.05$). The metal(loid) concentrations in mosses from SKS Mt. were not statistically analyzed in this way because

Table 1 Recovery (%) for metal(loid)s in certified reference poplar leaves (GBW07604) and detection limits (μg/L)

	Recovery	Detection limits
Al	88 ± 2	0.2
As	104 ± 3	0.3
Ba	103 ± 2	0.05
Cd	104 ± 2	0.05
Cr	91 ± 2	0.05
Cu	97 ± 3	0.2
Fe	110 ± 5	0.4
Mn	107 ± 3	0.3
Mo	106 ± 4	0.05
Ni	98 ± 2	0.05
Pb	102 ± 3	0.05
Sb	91 ± 2	0.05
Sr	88 ± 2	0.05
Ti	98 ± 3	0.3
Zn	100 ± 4	0.3

these mosses were sampled at different elevations. Linear regression analysis was performed for the metal(loid) concentrations in mosses from different elevations of SKS Mt. Principle component analysis (PCA) was applied to the metal(loid) concentrations of the mosses from SKS Mt. to help categorize the metal(loid)s from different sources. PCA was conducted by applying Varimax Rotation with Kaiser Normalization.

Results and Discussion

The concentrations of metal(loid)s in mosses from representative sites and SKS Mt. are listed in Table 2. Among the four representative sites, Al concentrations of mosses were similar ($p > 0.05$) while other metal(loid) concentrations varied significantly ($p < 0.05$). The mosses from CS site exhibit low concentrations of As, Cd, Cu, Ni, Pb, Sb, Sr, and Zn. The mosses from ISW site were characterized by high concentrations of As, Cr, Cu, Fe, Mn, Mo, Pb, and Zn. The mosses from AP site were characterized by high concentrations of As, Ni, Sb, and Zn. The mosses from BR site were characterized by high concentrations of As, Ba, Cd, Cu, Mn, Pb, Sb, Sr, and Ti. Regarding the mosses from SKS Mt., the concentrations of Al, Fe, and Ti were similar to those from CS site. The concentrations of other elements were typically within the range of metal (loid) concentrations of the representative sites. This may indicate that the metal(loid) concentrations of the mosses from SKS Mt. result from influences of the representative metal(loid) sources in Guiyang.

Metal(loid) concentrations of mosses from SKS Mt. were compared with those from other places that were monitored using the same type of moss (Table 2). The concentrations of Cd, Cr, Cu, Fe, Mn, and Zn in mosses from SKS Mt. were notably higher than those of the mosses from Gongga Mountain (Liang et al. 2008), a remote site in southwest China. This result indicates considerable atmospheric deposition of metal(loid)s in Guiyang. In comparison with mosses from Sheshan Mt. of Shanghai (Cao et al. 2008), the largest city in China, mosses from SKS Mt. showed higher concentrations of Cd, Cr, Cu, and Pb, but lower concentration of Zn. This indicates the high atmospheric metal(loid) pollution in Guiyang, although this city is much smaller than Shanghai.

Variations in the concentrations of metal(loid)s at different elevations of SKS Mt. are shown in Fig. 2. To quantify the variation, the concentration gradient (CG) of an element is calculated as the coefficient of linear regression of elemental concentration versus elevation. The concentrations of Mo and Zn are not suitable for calculation of CGs because p is >0.05 for them. CG < 0 means a decline trend of the elemental concentration with elevation, whereas CG > 0 means an opposite trend. The concentration gradients were ranked Fe < Al < Ti < Ba < Mn < Sr < Cr < Cu < Ni < Cd, Sb < As < Pb (Table 2). Fe, Al, and Ti in the mosses exhibited the most negative CGs (−29.7, −24, and −4.6, respectively), indicating a strong decline in atmospheric deposition of these elements with elevation. The concentration of As exhibits a relatively even distribution, and that of Pb shows a slight increase with elevation. Similarly, previous studies have reported the increase of moss Pb with rising altitude in the Alps (Zechmeister 1995; Gerdol and Bragazza 2006) and the Tatra Mountains in Slovakia (Soltes 1998).

To understand the causes for the different variations of metal(loid) concentrations with elevation, it is important to know the sources of elements. To evaluate the sources of metal(loid)s in mosses, it is necessary to normalize raw concentrations in mosses relative to soil or crustal abundance by calculating enrichment factors (EFs) (Bargagli et al. 1999). The reference element is considered to be an abundant element in the earth's crust (e.g., Al, Fe, Ti). Ti was used as the reference element and the EF was calculated as:

$$EF = [\mathrm{Element/Ti}]_{\mathrm{moss}} / [\mathrm{Element/Ti}]_{\mathrm{soil}}$$

A low EF value (approximately 1) indicates that the element in the moss is largely derived from crustal material; a median EF (2–10) indicates that in addition to the soil source, the element in the moss is partially derived from anthropogenic sources; and a high EF (>10) indicates that the element in the moss is largely derived from anthropogenic sources (Aničić et al. 2007).

Table 2 Metal(loid) concentrations (mg/kg) in moss (*H. microphyllum*) and soil from Guiyang City, China

Locations	Al	As	Ba	Cd	Cr	Cu	Fe	Mn	Mo	Ni	Pb	Sb	Sr	Ti	Zn
CS (n = 5)															
Min	1050	0.02	29.4	0.6	31.6	9.0	646	52.7	0.6	1.0	3.9	0.5	19.8	650	38.1
Max	7216	1.4	77.9	1.2	150	16.7	4043	137	0.8	3.9	10.5	0.7	30.7	174	140
Mean	3469[a]	0.4[b]	51.3[b]	0.8[b]	73.6[abc]	13.7[b]	2141[b]	94.0[c]	0.7[b]	2.1[b]	6.1[c]	0.6[b]	25.5[c]	358[b]	67.6[c]
SD	2270	0.7	20.2	0.3	134	3.4	1322	34.4	0.1	1.9	2.7	0.1	4.9	194	41.9
ISW (n = 5)															
Min	2573	4.6	59.1	1.7	57.3	23.2	4495	486	2.3	1.5	98.3	2.6	64.1	368	182
Max	6483	9.5	124	3.4	99.3	43.4	6307	2076	5.8	4.7	354	3.8	79.4	782	513
Mean	3579[a]	6.1[a]	88.6[ab]	1.8[a]	89.0[a]	35.3[a]	5046[a]	1354[a]	3.9[a]	2.8[b]	181[a]	3.2[a]	69.2[b]	472[a]	352[a]
SD	1645	1.9	23.5	0.5	20.0	7.6	757	599	1.4	1.3	101	0.5	6.5	175	129
AP (n = 4)															
Min	3721	4.9	51.8	1.0	5.9	18.2	1055	40.9	0.1	23.3	17.7	7.4	33.6	128	184
Max	3994	8.6	73.7	1.2	17.1	26.5	1770	63.1	0.6	28.5	26.8	10.6	37.7	239	432
Mean	3880[a]	6.3[a]	62.0[ab]	1.1[b]	9.9[b]	22.3[b]	1375[b]	49.0[c]	0.3[b]	26.7[a]	21.6[b]	8.7[a]	36.3[bc]	175[b]	268[a]
SD	141	2.0	11.0	0.1	6.3	4.1	363	12.2	0.4	3.0	4.7	1.7	2.3	58	142
BR (n = 5)															
Min	2173	3.8	59.6	2.3	21.7	24.3	1529	133	1.5	1.9	49.1	3.2	80.0	224	91.8
Max	5088	10.9	180	3.8	44.9	46.8	4574	333	2.3	5.0	115	6.4	140	613	153
Mean	3717[a]	8.3[a]	121[a]	3.1[a]	32.4[c]	37.7[a]	3201[ab]	249[b]	1.8[a]	3.5[b]	86.3[a]	5.1[a]	118[a]	433[a]	122[b]
SD	1422	2.8	46	0.6	9.2	8.5	1371	82	0.4	1.2	25.9	1.3	25	157	27
SKS Mt. (n = 29)															
Min	496	2.0	19.0	0.7	4.0	5.6	500	61.7	0.4	1.5	32.2	0.9	18.8	48.8	99
Max	8849	8.5	186	4.5	60.3	38.9	10,637	251	3.3	16.6	133	5.5	115	1651	362
Mean	3007	5.6	88.8	2.4	24.3	20.9	3390	159	2.0	6.3	73.2	3.1	53.5	406	205
SD	2156	2.0	49.3	1.0	15.6	8.9	2614	52	0.9	4.2	22.9	1.2	26.4	415	66
CG	−24.0	0.002	−0.61	−0.01	−0.19	−0.13	−29.7	−0.52		−0.05	0.17	−0.01	−0.37	−4.61	
Mean EF	0.9	13.2	8.7	558	5.7	18.8	1.5	7.1	20.9	3.8	83.4	29.8	21.8		74.4
Gongga Mt.*				0.1	0.2	0.3	33.2	3.2		0.2	1.8				4.0
Shanghai**				1.28	2.52	13.6					16.8				489
Guizhou Soil***	61,600	13.3	215	0.133	86.6	25.7	41,700	591	1.8	33.7	29.3	1.85	66.9	6200	82.4

n, number of samples; SD, standard deviation; CG, the concentration gradient of an element in mosses with rising elevation; EF, enrichment factor

* Liang et al. (2008); ** Sheshan Mt. in Shanghai (Cao et al. 2008); *** Wang and Wei (1995)

Difference between four representative metal(loid) source sites are indicated with different letters in superscript (a, b, c)

Al and Fe in SKS Mt. mosses had low EF values (0.93 and 1.5), indicating that Al and Fe were primarily derived from soil. The soil origin of Al, Fe, and Ti in mosses has been demonstrated in previous studies (Steinnes 1995; Gerdol et al. 2002). This is consistent with the fact that the concentrations of Al, Fe, and Ti in the mosses notably declined with elevation (Fig. 2), because the influence of soil dust is stronger at lower elevations and is weaker at higher elevations.

The EF values of Ba, Cr, Mn, and Ni in the SKS Mt. mosses are in the median level of 8.7, 5.7, 7.1, and 3.8, respectively (Table 2), indicating that in addition to a soil source, Ni, Cr, Mn, and Ba are partially contributed by anthropogenic sources. Ni can be partially derived from the AP site, because the concentrations of Ni in the mosses are highest at this site. The anthropogenic portion of Cr and Mn can be attributed to ISW where moss Cr and Mn concentrations are highest among the representative sites. The anthropogenic portion of Ba is possibly derived from road traffic because the mosses at the BR site show a high Ba concentration of 121 mg/kg. Brake wear is likely the main source of Ba because Ba is often used in the form of barite in the filler material of brake pads (Gietl et al. 2010).

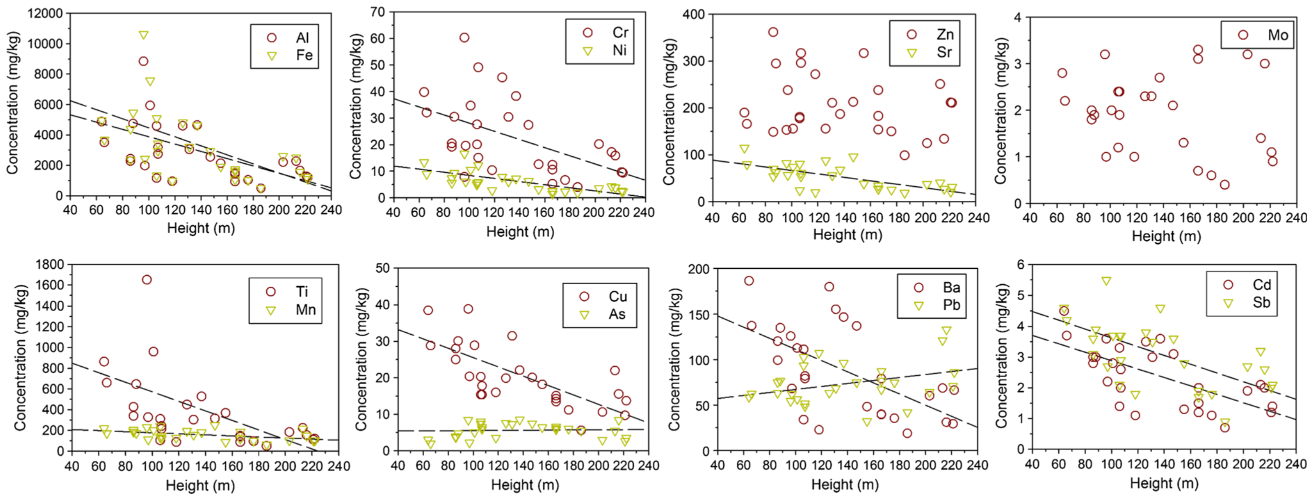

Fig. 2 The distribution of the metal(loid) concentrations of the mosses with increasing elevation (the *dashed lines* represent the linear regression of data)

The notably high EF values for As, Cd, Cu, Mo, Pb, Sb, Sr, and Zn (13.2, 558, 18.8, 20.9, 83.4, 29.8, 21.8, and 74.4, respectively) in the SKS Mt. mosses indicate that these elements are predominantly derived from anthropogenic sources. As mentioned above, the mosses from BR, ISW, and AP sites show high concentrations of metal(loid)s (Table 2), so the road traffic and the metal smelting processes are considered the dominant sources for these elements. Note that Cd and Cu can originate from tire wear, and Cu, Mo and Sb can originate from brake wear (Hjortenkrans et al. 2007). Strontium was commonly reported to derive from natural sources (Bačeva et al. 2013). In this study the high EF of Sr and the high Sr concentration of the mosses from BH site indicate that Sr can be highly derived from anthropogenic sources. Srimuruganandam and Shiva Nagendra (2011) reported that Sr in aerosols can originate from brake and tire wear. Antimony can also be derived from brake wear and also the emissions of AP. Arsenic and Zn in the SKS Mt. mosses can be attributed to ISW and AP, because the mosses from these two sites show high As and Zn concentrations (Table 2). The mosses from ISW site showed the highest Pb concentrations, indicating that Pb may originate from high temperature processes in ISW, because As and Pb were commonly found to be emitted from high temperature processes in industry (Gao et al. 2002).

The PCA results are shown in Table 3. Three factors were separated with eigenvalues >1, explaining a sum of 83.9 % of the total variance. Factor 1 was characterized primarily by Al, Ba, Cd, Cr, Cu, Fe, Mn, Mo, Ni, Sb, Sr, and Ti. This factor explains 61.6 % of the total variance. As mentioned above, Al, Fe, and Ti are predominantly derived from soil; Cr, Mn and Ni are partially derived from soil; and Ba, Cd, Cu, Sb, and Sr are derived from vehicle

Table 3 Rotated component matrix for data of the moss *H. microphyllum* on the SKS Mt.

Element	Factor 1	Factor 2	Factor 3
Al	**0.94**	0.01	−0.19
As	0.10	**0.65**	0.02
Ba	**0.87**	0.13	0.12
Cd	**0.96**	0.11	0.01
Cr	**0.91**	0.07	−0.12
Cu	**0.89**	−0.01	0.03
Fe	**0.91**	−0.02	−0.22
Mn	**0.73**	**0.51**	0.18
Mo	**0.56**	0.38	−0.38
Ni	**0.93**	−0.13	−0.04
Pb	−0.22	**0.75**	−0.25
Sb	**0.92**	0.19	−0.01
Sr	**0.89**	0.05	0.25
Ti	**0.89**	−0.18	−0.20
Zn	−0.03	−0.11	**0.91**
Eigenvalue	9.24	1.53	1.29
% Variance	61.6	10.2	8.58

PCA loadings >0.5 are shown in bold

and road wear. The metal(loid)s associated with vehicle and road wear tend to accumulate in road dusts (Hussein et al. 2008). Therefore, Factor 1 may be interpreted as representing the resuspension of road dusts that consist of soil particles and traffic-related particles. Factor 2 was loaded primarily by Mn, As, and Pb. Manganese is included in both Factor 1 and Factor 2 because Mn can be derived from both soil and ISW. Arsenic and Pb in atmosphere are usually related to emissions from high temperature processes in industry (Gao et al. 2002). Thus Factor 2 was considered to have resulted from high temperature processes. Factor 3 was characterized by only Zn. Zn and some other metal(loid)s are derived from the same sources (ISW and AP), but only Zn is included in this factor. This is possibly because the uptake of Zn in the moss tissues is closely related to the moss metabolism (Boquete et al. 2014).

This study revealed that mosses at different elevations may serve as indicators of altitudinal variation of atmospheric metal(loid) deposition. The metal(loid)s in the atmosphere of Guiyang are predominantly affected by soil, metal smelting processes and traffic-related road dusts. The altitudinal deposition of soil-originated metals differs largely from that of anthropogenic metal(loid)s. As indicated by the moss metal(loid) concentrations, the soil-related elements tend to deposit at lower elevations and their deposition at higher elevations is less; whereas the atmospheric deposition of elements from anthropogenic sources does not vary largely with elevation.

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