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The measurement of ambient air moisture stable isotope composition for the accurate estimation of evaporative losses



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ABSTRACT

Accurate estimation of evaporative losses from a water body, using the Craig-Gordon model and the stable hydrogen and oxygen isotope composition of water, requires knowledge of the stable isotope composition of ambient air moisture. This is rarely measured in the field, and it is usually estimated assuming that recent rainfall remains in isotopic equilibrium with atmospheric moisture. However, the ambient air moisture stable isotope composition may vary significantly at different heights above the water body.

In this study, we set up outdoor pan evaporation experiments and simultaneously measured the stable isotope composition of ambient moisture in the atmosphere at three different heights. Using these measurements, we calculated evaporative losses, compared them with the observed losses in the pan, and assessed the uncertainty introduced by differences in ambient moisture measurements.

Three main steps in the experimental method:

- Daily water sampling from the evaporation pan for analysis of stable hydrogen and oxygen isotope compositions.
- Recording the stable isotope composition of ambient air moisture at three different heights using the Picarro L2130-i system over a period of experiments.
- Calculating evaporative losses from the pan using the Craig-Gordon model and ambient air stable isotope composition measured at three different levels and comparing to the observed losses.

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Method details

This method describes a technical and conceptual setup for measuring the stable hydrogen and oxygen isotope compositions of ambient air for evaporation pan experiments. The differences recorded at the three levels above the ground during different weather conditions were used to calculate water fraction losses and to compare them with the losses observed in the pan. The uncertainty arising from measuring ambient air at different levels was used to evaluate the overall accuracy of the evaporation calculation method using the Craig-Gordon model.

Experimental design

Two pan evaporation experiments were conducted in March and April 2021 at The University of Western Australia (UWA), Perth, Western Australia. The galvanised-iron and metal-coloured standardised class-A evaporation pan (diameter 1.2 m, depth of 0.25 m, www.bom.gov.au) was positioned outdoors on a hard plastic pallet (16 cm thick) next to the UWA weather station (32.0 °S, 115.8 °E, altitude 10 m). Short grass (~5 cm) covered the area around the pan. Local municipal tap water was used for each experiment (TDS < 800 mg/L). The daily evaporation rate was calculated by gauging the water level with a ruler between 8.30 and 9.00 AM every day, with 1 mm resolution. Measured water levels were recorded, and 2 mL water samples were collected from the pan with a disposable pipet and stored in a glass vial for stable isotope analysis (Thermo Fisher Scientific Inc., London, England). The collected samples corresponded to <0.01% of the water volume in the pan, so it was negligible for evaporative loss calculation. Additionally, a standardised rainwater collector preventing evaporative losses, as designed at International Atomic Energy Agency by Gröning (produced by Palmex Ltd, Zagreb, Croatia), was installed at the pan to monitor the stable isotope composition of potential precipitation [1].

Air and water temperature, and air relative humidity were recorded using *in-situ* data loggers. Onset Hobo H08–32–08 data loggers (Onset, Bourne, MA, USA) were used for air temperature recording with \pm 0.02 °C resolution and relative humidity with \pm 3% resolution at 10 cm above the pan. Additionally, Hobo U12–014 Thermocouples (\pm 0.32 °C resolution) were installed at the edge of the pan to record the water temperature. Solar radiation and wind speed data were taken from the UWA Weather Station located 15 metres away from the pan. Meteorological data were recorded at 15-min intervals and used to calculate daily means for periods between samplings. Solar radiation was recorded using a Middleton EQ08-E Solar Radiation sensor (Middleton Solar, North Geelong, VIC, Australia).

Three 10 m long perfluoro alkoxy alkane (PFA) tubes (1.2 mm wall, 6.35 mm outside diameter, Thermo Fisher Scientific Inc, London, England) were attached to collect atmospheric air from different heights. The first tube was installed just above the ground over grass at the pan. The second tube was installed on top of the pan ~5 cm above water level. The third tube was installed 3 m above the ground. The air was continuously pumped through all tubings with a constant flow rate of 70 mL/min. The tubes were connected via Vici Multiposition Microelectric Valve Actuators (VICI Valco Instruments Co. Inc., Houston, TX, USA) to an Isotope and Gas Concentration Analyser to Picarro L2130-i (Picarro, Santa Clara, CA, USA) to continuously analyse the stable isotope composition of ambient air moisture. The ambient air moisture stable isotope composition was measured at all three heights every second for 10-min intervals from each pipe, starting from the lowest to the highest level, and analysed by Picarro L2130-i water vapour analyser. The first five minutes of each measurement period for the isotopic composition of ambient air moisture were removed to allow purging the system and to reduce the instrument memory effect. The rest of the measurements were averaged arithmetically. The liquid laboratory standards used for normalisation were measured at 24 h intervals. The isotope composition of pan water was also analysed using the same system, Picarro L2130-i equipped with a V1102-I vaporiser.

All results were reported as 1000 of isotope delta (δ) in permille (∞) on the VSMOW2-SLAP2 international stable isotope scale after multipoint normalisation and drift correction. The raw vapour data (δ^2 H and δ^{18} O) were normalized using a two-point normalization based on two laboratory standards (Polish spring water, POL δ^2 H= $-61.74 \ \infty$, δ^{18} O= $-8.14 \ \infty$; Canadian spring water, CAD δ^2 H= $-135.50 \ \infty$, δ^{18} O= $-17.94 \ \infty$). The third standard was added for normalisation of liquid water samples to increase precision (Perth Ocean water, POW δ^2 H = $11.66 \ \infty$, δ^{18} O = $3.92 \ \infty$). These laboratory standards were calibrated using primary reference materials VSMOW2 and SLAP2 provided by the International Atomic Energy Agency.

The stable isotope composition of local tap water used in the two experiments was slightly different (Exp. 1 $\delta^2 H = -0.06 \%$, $\delta^{18}O = 0.34 \%$, and Exp 2. $\delta^2 H = 14.02$ and $\delta^{18}O = 3.64 \%$). This difference results from variability in Perth municipal water that reflex mixing of three water sources in the water supply (groundwater, surface dam water and desalinated water). These values are far below the maximum enrichment level, and changes in the stable isotope composition over the evaporation observation period well reflected the progress of evaporation observed in the pan.

Variables and equations

The evaporative loss calculations were made using HydroCalculator, an open source software accessible at http://hydrocalculator.gskrzypek.com [2]. The software was designed to calculate evaporative loss based on the modified Craig–Gordon model. The Craig-Gordon model calculations are based on the relative change of the stable isotope composition in the pan (between initial pan water δ_p and final δ_L) and requires a few additional parameters from those measured [3]. These parameters are air temperature (T), relative humidity (h), and the stable isotope composition of ambient air moisture (δ_A). In the experiments described below we report the mean calculated value of evaporative loss using measurements of both δ^{18} O and δ^{2} H.

Air temperature (T) and relative humidity (h)

The stable isotope composition of water in the pan, and the progress of evaporation during experiments can be affected by the relative humidity (h) and temperature (T) of air. Therefore, it is best to measure the parameters locally, directly at the place where evaporation occurs.

Stable isotope composition of water (δ_P , δ_L)

Under non-steady state conditions (no water added, water level decreases as evaporation progresses), the evaporation losses are reflected in the increase of the stable isotope delta values of the water remaining in the pan. The stable isotope composition of water progressively changes from the initial value δ_P to the final value δ_L , if the stable isotope composition does not reach the maximum enrichment level in heavy isotopes which depend on the weather conditions.

Stable isotope composition of ambient air moisture (δ_A)

The stable isotope signature of ambient air moisture (δ_A) influences the stable isotope composition of δ_L and therefore it needs to be known to calculate evaporative losses. However, δ_A can vary temporally and spatially [4,5] because of the difference in originated air masses, time of the day and vertical mixing, and evapotranspiration [6]. There are three ways to obtain a stable isotopic composition of ambient air moisture (δ_A):

- the direct measurement on-site using, e.g., the Picarro instrument (as in this experiment);
- analysing the isotopic composition of local precipitation and calculating δ_A assuming that the moisture in the atmosphere is in isotopic equilibrium with recent precipitation;
- analysing the local precipitation isotope composition, calculating δ_A and then correcting it by applying a local evaporation line (LEL).

Although it is logistically challenging in distant areas, direct measurement of the isotopes of ambient air is the most precise method if conducted over the entire observation period [2,6].

Non-steady state model evaporation calculation

In the non-steady state model, the volume of the water and its isotope composition change solely because of evaporative losses. The evaporative loss, as the fraction (f) of the initial volume of water, can be formulated as the following equation (Eq. (1)):

$$f = 1 - \left[\frac{(\delta_L - \delta^*)}{(\delta_P - \delta^*)}\right]^{\frac{1}{m}}$$
(1)

where δ_L (sampling 1) is the initial isotope composition, and δ_P is its final value of the water body (sampling 2). δ^* is the limiting isotopic composition of the water body [7] and is expressed by the following equation (Eq. (2)):

$$\delta^* = \frac{h\delta_A + \epsilon_K + \frac{\epsilon}{a^+}}{h - 10^{-3} \left(\epsilon_K + \frac{\epsilon^+}{a^+}\right)} \tag{2}$$

and m is defined as (Eq. (3)):

$$m = \frac{h - 10^{-3} \left(\frac{\epsilon^{+}}{\alpha^{+}} + \epsilon_{K}\right)}{1 - h + 10^{-3} \epsilon_{K}}$$
(3)

where h is relative humidity as fraction, ϵ^+ is an equilibrium isotope fractionation factor depending on temperature, and is represented as (Eq. (4)):

$$\varepsilon^+ = (\alpha^+ - 1) \times 1000 \tag{4}$$

According to Horita and Wesolowski [8] the equilibrium isotopic fractionation for water (a^+) between the liquid and vapour phases is governed primarily by temperature (in Kelvin degrees), and the relation is as follows (Eqs. (5) and (6) [3]).

$$10^{3} \times \ln \alpha^{+} = 1158.8 \times T^{3} \times 10^{-9} - 1620.1 \times T^{2} \times 10^{-6} + 794.84 \times T \times 10^{-3} - 161.04 + 2.9992 \times 10^{9} \times T^{-3}$$
(5)

$$10^{3} \times \ln \alpha^{+} = -7.685 + 6.7123 \times T^{-1} \times 10^{3} - 1.6664 \times T^{-2} \times 10^{6} + 0.35041 \times 10^{9} \times T^{-3}$$
(6)

Total isotopic fractionation (ϵ) consists of temperature-dependant equilibrium isotope fractionation (ϵ^+) and kinetic isotope effects ($\epsilon_{\rm K}$), calculated as per Eq. (7) [9]

$$\varepsilon = \varepsilon^+ + \varepsilon_K \tag{7}$$

The kinetic isotope fractionation $\varepsilon_{\rm K}$ is expressed as follows:

$$\varepsilon_K = (1-h) \times C_K \tag{8}$$

where C_K is the kinetic fractionation and experimentally determined as 12.5 for hydrogen and 14.2 for oxygen by Gonfiantini [9].



Fig. 1. The comparison between the calculated and measured evaporative loss at the ground, the pan, and 3 m at height for Experiment 1.

Experiment #1

Two pan evaporation experiments were conducted (March and April 2021) over 10-day periods each. The water evaporative loss calculations for each experiment were repeated three times using the stable isotope composition of ambient moisture from the three monitored heights. The calculated losses were compared with the losses observed in the pan, based on the water level changes.

For the first experiment, the pan was filled with 230.7 L of local tap water to a level of 204 mm. The evaporation caused the water level to decrease by about 3–4 mm per day. At the end of the experiment, 15.2% of the volume was lost. In the pan, 84.8% of the initial volume was left ~196 L (Tables 1 and 2). The daily mean temperature during the observation period varied between 19.27 and 21.82 °C and the relative humidity between 46.0 and 62.5%.

The stable isotope composition of water became progressively more positive during each day, changing from δ^{18} O 3.64 ‰, δ^{2} H 14.02 ‰ at the start to δ^{18} O 5.71 ‰, δ^{2} H 23.07 ‰ at the end of the experiment 10 days later (Table 1).

The ambient moisture stable isotope composition varied according to the day of sampling and the sampling height. The temporal variation was within 1.6 ‰ for δ^{18} O and 7.9 ‰ for δ^{2} H. The differences recorded on the same day at different height levels were up to 1.2 ‰ for δ^{18} O and 9.4 ‰ for δ^{2} H. Consequently, the losses calculated using ambient air stable isotope composition from the three levels were different, and the difference increased, cumulating over the time of the experiment (Table 2). On day 10, the difference between the observed in pan total evaporative loss and the total loss calculated using ambient moisture signature at the ground level was 0.4%, at the pan level was 1.7% and at 3 m height 3.3%. However, the correlations between the observed loss and calculated loss using ambient air from different levels were strong and statistically significant (R²=0.99, *p*<0.01) but their slopes were different (Fig. 1).

Experiment #2

In the second experiment, the pan was filled with 124.4 L, and the initial water level was 220 mm. The water level decreased by an average of 2 mm per day (Table 3). The mean daily temperature was similar to those observed during the first experiment, varying between 19.24 and 20.22 °C, but relative humidity was much higher, varying between 70.1 and 77.1%.

During the second experiment, on the seventh day (30/04/2021) a short 8 mm rainfall occurred, adding water to the pan. The stable isotope composition of the pan water and its volume was corrected using the volume and the stable isotope composition of the added rain (δ^2 H = -10.72 ‰, δ^{18} O =-3.18 ‰) sampled by the rainwater collector. The stable isotope composition of water changed during the second experiment much less than during the first experiments between δ^{18} O =-0.34 ‰, δ^2 H =-0.06 ‰ at the start, and

Table 1 First pan experiment, stable isotope results, and weather parameters.

Date/time	time Water isotopes in the pan $\delta^{18}O$ (% $\delta^{2}H$ (% VSMOW) VSMOW)		Water level (mm)	Water volume in pan (dm ³)	Field measured water volume loss from start (%)	Temperature (°C), mean from start	Relative Humidity (%), mean from start	Solar radiation (W/m ²), mean from start	Wind speed (m/s), mean from start
23/03/2021 08.30 Start	3.64	14.02	204	230.7	0	-	-	-	-
24/03/2021 08.30	4.01	15.75	201	227.3	1.5	19.44	46.0	199.46	4.36
25/03/2021 08.30	4.27	16.90	198	223.9	2.9	19.27	52.5	202.32	4.12
26/03/2021 08.30	4.55	18.09	195	220.5	4.4	19.47	54.9	202.37	3.82
27/03/2021 08.30	4.73	18.74	190	214.9	6.9	19.79	56.9	201.68	3.69
28/03/2021 08.30	4.92	18.92	188	212.6	7.8	20.05	58.2	201.51	3.51
29/03/2021 08.30	5.23	20.45	184	208.1	9.8	20.54	58.0	203.08	3.31
30/03/2021 08.30	5.50	21.65	178	201.3	12.7	21.34	58.4	203.08	3.24
31/03/2021 08.30	5.72	22.57	177	200.2	13.2	21.66	60.9	203.08	3.26
01/04/2021 08.30 End	5.71	23.07	173	195.7	15.2	21.82	62.5	203.08	3.23

Table 2

The isotope composition of ambient air moisture during the first experiment at the ground level, pan level, and 3 m at height and calculated evaporative loss from the pan.

Date	Vapour at the ground level			Vapour at pan level			Vapour at 3 m height		
	δ^{18} O (%0)	δ^2 H (%0)	Loss (%)	δ ¹⁸ Ο (‰)	$\delta^2 \mathrm{H}$ (‰)	Loss (%)	δ^{18} O (%0)	$\delta^2 \mathrm{H}$ (%0)	Loss (%)
23.03.2021	-	-	-	-	-	-	-	-	-
24.03.2021	-12.57	-85.92	2.2	-13.51	-91.23	2.3	-13.80	-95.32	2.4
25.03.2021	-12.58	-87.54	4.0	-13.50	-92.89	4.4	-13.69	-95.36	4.5
26.03.2021	-12.54	-87.43	6.1	-13.44	-92.83	6.6	-13.69	-95.26	6.9
27.03.2021	-12.49	-87.86	7.5	-13.25	-92.46	8.2	-13.59	-95.34	8.6
28.03.2021	-12.46	-88.24	8.6	-13.15	-92.45	9.4	-13.46	-94.99	9.9
29.03.2021	-12.12	-86.28	10.7	-12.79	-90.18	11.6	-13.22	-93.81	12.4
30.03.2021	-11.66	-84.09	12.5	-12.35	-87.88	13.5	-12.76	-91.57	14.5
31.03.2021	-11.43	-82.82	14.7	-12.06	-86.25	15.9	-12.47	-89.89	17.3
1.04.2021	-11.25	-81.94	15.6	-11.80	-85.02	16.9	-12.23	-88.59	18.5



Fig. 2. The comparison between the calculated and measured evaporative loss at the ground, the pan, and 3 m at height for Experiment 2.

 δ^{18} O 1.00 ‰, δ^{2} H 5.47 ‰ at the end 10 days later (Table 1). The addition of rainfall on 30 April decreased δ^{18} O by 0.25 ‰ and δ^{2} H 0.94 ‰, but this difference is accounted for in evaporation calculations.

The stable isotope composition of ambient moisture varied less during the second experiment than during the first experiment, and the temporal variation was within 1.9 % for δ^{18} O and 12.6 % for δ^{2} H. However, the differences recorded on the same day at different height levels were greater, with δ^{18} O variation within 2.1 % and δ^{2} H variation within 9.4 %. The overall calculated evaporative losses were lower (5.3%, 6.9% and 6.7%, Table 4) compared to the first experiment (15.6%, 16.9% and 18.5%), reflecting lower evaporation during a more humid period. The overall differences between the measured and calculated loss for the 10-day period were lower when compared to the first experiment and were 2% at the ground level, 0.4% at the pan level and, 0.6% at 3 m level (Fig. 2).

The daily mean δ_A values at different heights varied and therefore contributed to uncertainty in evaporative loss calculations. The difference in calculated evaporative losses was up to 2.9% during Experiment 1 and up to 1.6% during Experiment 2, for 10-day observation periods. For Experiment 1, the difference between observed (15.2%) and calculated evaporative loss was the smallest when δ_A from the ground level was used (15.6%). This may suggest that during relatively dry conditions (RH 46 to 62.5%), evapotranspiration and evaporation from soil had a large influence on the moisture composition during evaporation from the pan. In Experiment 1, the differences led to more than 1% evaporation loss difference in calculations. During Experiment 2 the difference in ambient air stable isotope composition at difference in calculations. During Experiment 2, the relative humidity in the atmosphere was higher (RH 70.1 to 77.1%), and evaporation from soil or evapotranspiration contributed much less to the stable isotope composition of the atmosphere at the pan than in dry conditions.

Table 3 Second pan experiment, stable isotope results, and weather parameters.

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Date/time	Water isotopes in the pan δ^{18} O (‰ δ^{2} H (%0 VSMOW) VSMOW)		Water level (mm)	Water volume in pan (dm ³)	Field measured water volume loss from start (%)	Temperature (°C), mean from start	Relative Humidity (%),mean from start	Solar radiation (W/m ²), mean from start	Wind speed (m/s), mean from start
23.04.2021 09:00 Start	-0.34	-0.06	220	124.4	-	-	-	-	-
24.04.2021 09:00	-0.14	0.87	218	123.3	0.9	19.87	70.1	262.9	2.03
25.04.2021 09:00	0.05	1.65	217	122.7	1.4	20.02	74.2	262.9	1.91
26.04.2021 09:00	0.24	2.29	216	122.1	1.8	19.68	77.1	262.9	1.84
27.04.2021 09:00	0.43	3.06	214	121.0	2.7	19.99	75.5	262.9	1.77
28.04.2021 09:00	0.57	3.69	212	119.9	3.6	20.00	76.5	262.9	1.71
29.04.2021 09:00	0.75	4.41	210	118.8	4.5	20.22	76.4	264.9	1.74
30.04.2021 09:00	0.74	4.31	210	118.8	4.5	20.01	77.1	266.9	2.33
1.05.2021 09:00	0.77	4.99	206	116.5	6.4	19.64	75.9	268.4	2.29
2.05.2021 09:00 End	1.00	5.47	204	115.4	7.3	19.24	75.6	269.5	2.28

Table 4

The isotope composition of ambient air moisture during the first experiment at the ground level, pan level, and 3 m at height and calculated evaporative loss from the pan.

Date Vapour at the ground level				Vapour at pa	n level		Vapour at 3 m height		
	δ^{18} O (%0)	δ^2 H (%0)	Loss (%)	δ^{18} O (%0)	δ^2 H (%0)	Loss (%)	δ^{18} O (%0)	δ^2 H (%0)	Loss (%)
23.04.2021	-	-	-	-	-	-	-	-	-
24.04.2021	-9.95	-75.50	0.9	-12.02	-88.65	1.2	-11.55	-87.90	1.2
25.04.2021	-9.15	-71.28	1.6	-11.10	-83.38	2.1	-10.74	-83.40	2.1
26.04.2021	-9.67	-73.80	2.4	-11.05	-82.77	3.1	-10.57	-81.08	2.9
27.04.2021	-9.29	-70.92	3.0	-10.83	-81.32	3.9	-10.46	-80.07	3.7
28.04.2021	-9.09	-69.93	3.5	-10.58	-79.70	4.5	-10.29	-78.96	4.4
29.04.2021	-8.66	-67.47	3.9	-10.12	-76.68	5.0	-9.75	-75.34	4.8
30.04.2021	-8.96	-68.72	3.6	-10.31	-77.16	4.5	-10.07	-76.74	4.4
1.05.2021	-9.28	-69.76	4.5	-10.71	-78.61	5.7	-10.47	-78.40	5.6
2.05.2021	-9.48	-70.73	5.3	-11.05	-80.20	6.9	-10.74	-79.64	6.7

Therefore, the observed evaporation (7.3%) was much closer to the calculated when δ_A from the pan level (6.9%) or the 3 m above the pan (6.7%) were used. This observation suggests change in boundary conditions and overall lower contribution from soil and vegetation water to the vapour at the study site during Experiment 2.

The small discrepancy between observed and calculated losses within $\pm 1.5\%$ still were observed and can be attributed to other environmental factors. For example, a wind velocity above 2 m/s may lift microdroplets from the water surface [10]. This mechnism is resulting in volume decrease in the pan but without stable isotope fractionation, whole droplets can be removed from the pan without evaporation. Therefore, the water remaining in the pan could have more negative delta avalues sugesting lower evaporation than actually observed [10]. Stronger winds disturbe aslo the equilibrium and laminar layers above the water surface, and hence a greater water loss occures.

During the experiments, weather parameters were recorded using *in-situ* data loggers charcterised by high precision and resolution ($T \pm 0.02$ °C and RH ±3%). However, even small changes in humidity and temperature can affect evaporation rate calculations. Therefore, to understend possible sources of uncertanity beyond variability in ambient moisture stable isotope composition, Kragten uncertanity analysis [11] was run for the range of variables observed in this study. The propagation of uncertanity in temperture measurments contributed to overall low sensitivity, and ± 1 °C difference led only up to 0.2% difference in evaporative loss calculations. The uncertanity in relative humidity had a higher impact on the final comabined uncertanity, and $\pm 5\%$ change in RH changed the calculation of evaporation by up to 5%. All these values are far below the discrepance between observed and calculated values that were recorded in our experiments (<3%).

Summary

Our observations suggest that measuring ambient air stable isotope composition in the field will improve the overall accuracy of the calculation and will help to understand boundary changes during different weather patterns. However, it is not critical if uncertainty n calculations of evaporative losses in the range of $\pm 1.50\%$ could be accepted. These results are valid for the range of parameters observed in our study conducted in mild Mediterranean climate, and it is advised to repeat these experiments for other studies if conducted under different climate conditions.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Cihat Adsiz: Investigation, Formal analysis, Writing – original draft. **Grzegorz Skrzypek:** Conceptualization, Methodology, Writing – review & editing, Supervision, Resources. **James McCallum:** Writing – review & editing, Supervision.

Data availability

Dataset is provided in supplemental materials.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.mex.2023.102265.

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