Divergent Aging Mechanisms of Calcium Arsenic Residue under Dry-Wet and Freeze-Thaw Cycles: Toxic Metal Mobility, Multiscale Physicochemical Characterization, and Escalated Ecological Risks

Xiaolong Zhao [1,2,†], Guangli Wang [1,2,†], Ying Du [1,2], Zhiying Zhao [1,2], Menghua Ran [1,2] and Dongyun Du [1,2,*]

[1] Hubei Province Engineering Research Center for Control and Treatment of Heavy Metal Pollution, College of Resources and Environmental Science, South-Central Minzu University, Wuhan 430074, China; 2022120861@mail.scuec.edu.cn (X.Z.); lgw4727@163.com (G.W.); 2022010048@mail.scuec.edu.cn (Y.D.); 1393881500@qq.com (Z.Z.); 2022110584@mail.scuec.edu.cn (M.R.)

[2] Key Laboratory of Catalysis and Energy Materials Chemistry of Ministry of Education, Wuhan 430074, China

* Corresponding author. E-mail: dydu666@mail.scuec.edu.cn (D.D.)

† These authors contributed equally to this work.

Table S1. Major elemental composition of CAR.

Element	Ca	As	Sx	Fe	F	Si	Mg	Zn	Al	Cu	Cd
Wt (%)	24.87	2.32	20.15	1.36	3.57	0.88	1.27	2.35	0.57	0.47	0.24

Table S2. Toxicity impact factors in the Potential Ecological Risk Index (I_{PER}) methodology.

Element	As	Zn	Cd
Toxicological response factor (T_r^i)	10	1	30

Table S3. Risk level evaluation of heavy metal pollution by E_r^i and $I_{PER.}$

E_r^i	Single potential ecological risk level	I_{PER}	Combined potential ecological risk level
＜40	Low ecological risk	＜150	Low risk
[40,80)	Moderate ecological risk	[150,300)	Moderate risk
[80,160)	High ecological risk	[300,600)	High risk
[160,320)	Considerable ecological risk	≥600	Severe risk
≥320	Extreme ecological risk		

Table S4. Potential Ecological Risk Values of CAR after DW and FT Cycles

Parameter (Grade, Contribution Rate)	Raw CAR	DW cycle 28 rounds	FT cycle28 rounds
E_{As}	8129.50 (Extreme, 79.47%)	8965.51 (Extreme, 80.14%)	8535.67 (Extreme, 80.19%)
E_{Zn}	100.51 (Strong, 0.98%)	125.66 (Strong, 1.12%)	118.13 (Strong, 1.11%)
E_{Cd}	1999.5 (Extreme, 19.55%)	2091.68 (Extreme, 18.74%)	2014.49 (Extreme, 18.7%)
I_{PER}	10229.51 (Severe Risk)	11187.85 (Severe Risk)	10668.29 (Severe Risk)

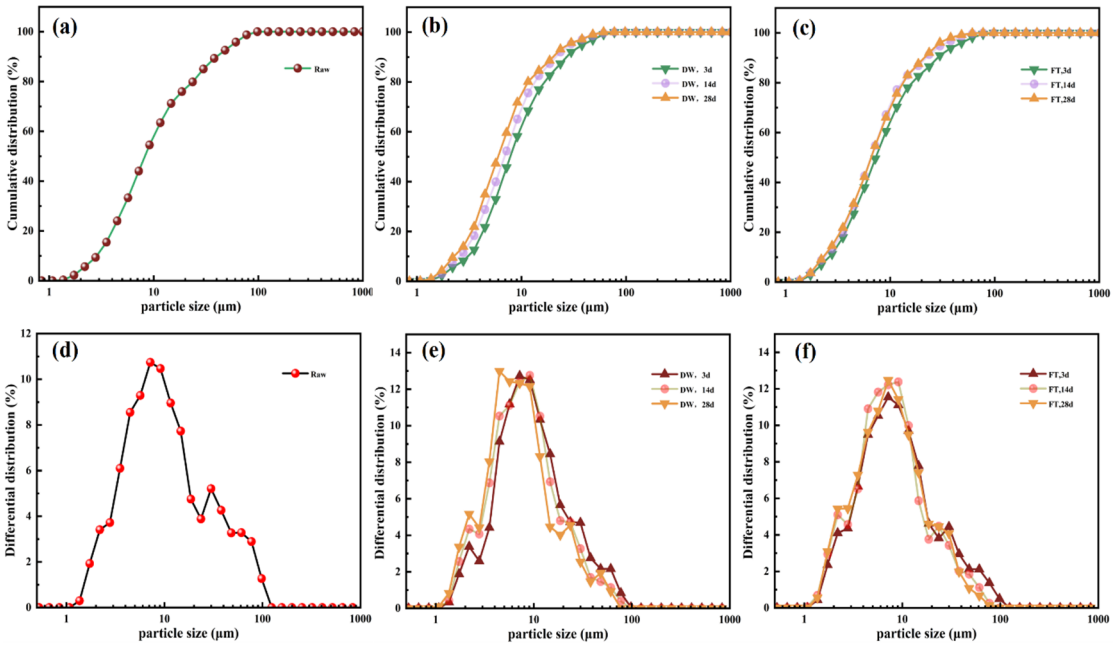

Figure S1. Presents the impacts of DW and FT on CAR particle size distribution and cumulative curves: (a, d) raw sample; (b, e) DW; (c, f) FT.

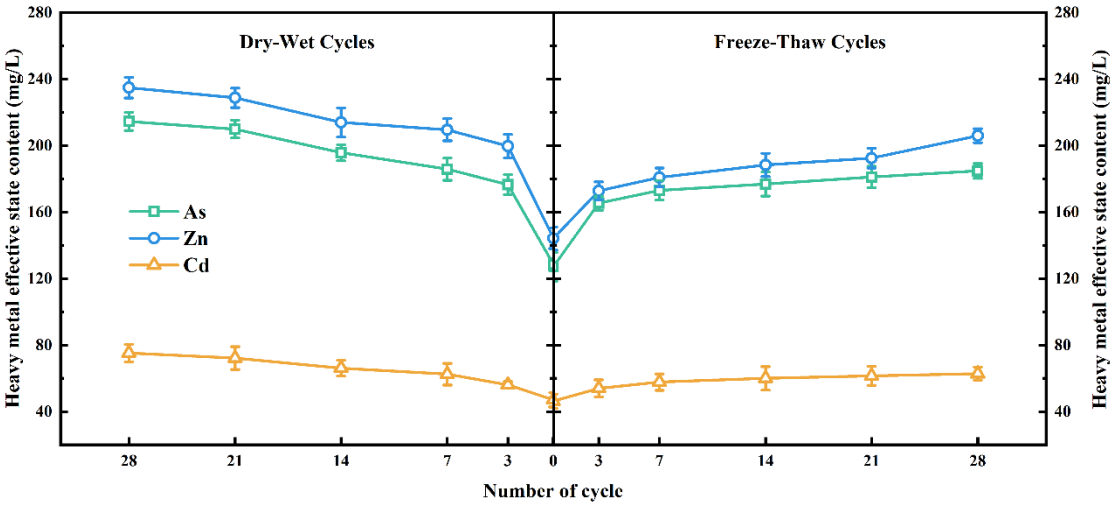

Figure S2. Variations in the concentrations of bioavailable As, Zn, and Cd in CAR after 1 to 28 cycles of DW and FT cycling.

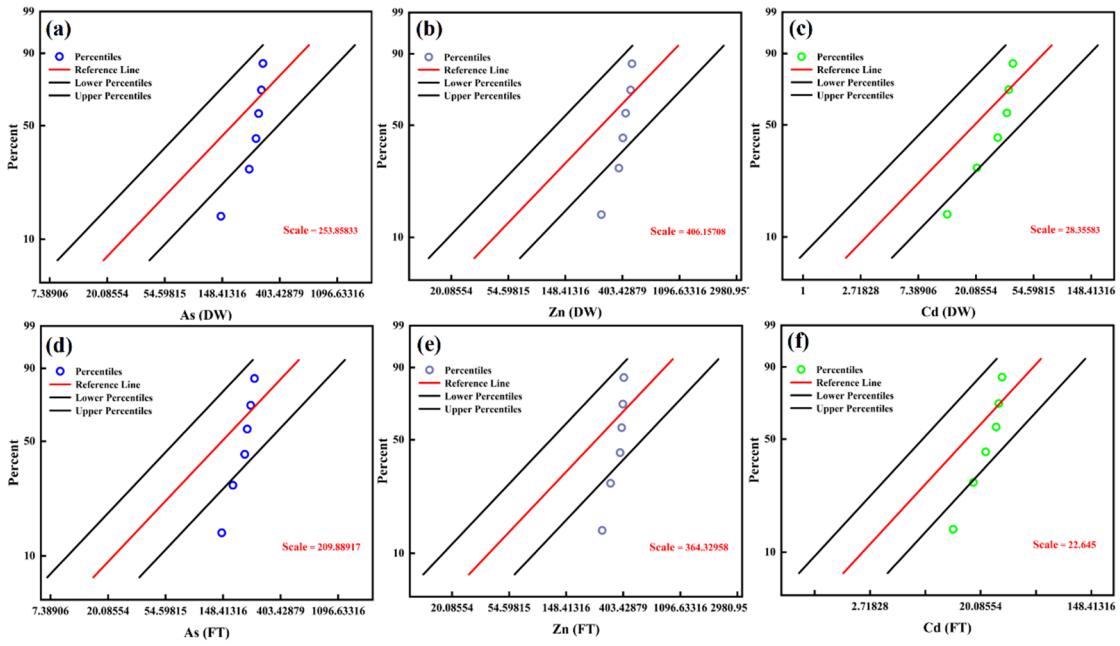

Figure S3. Exponential model of heavy metals in CAR under accelerated aging conditions for 28 years, subjected to DW and FT cycles. DW: (a) As, (b) Zn, (c) Cd; FT: (d) As, (e) Zn, (f) Cd.