Supporting Information for

Solid Phase Silver Sulfide Nanoparticles Contribute Significantly to Biotic Silver in Agricultural Systems

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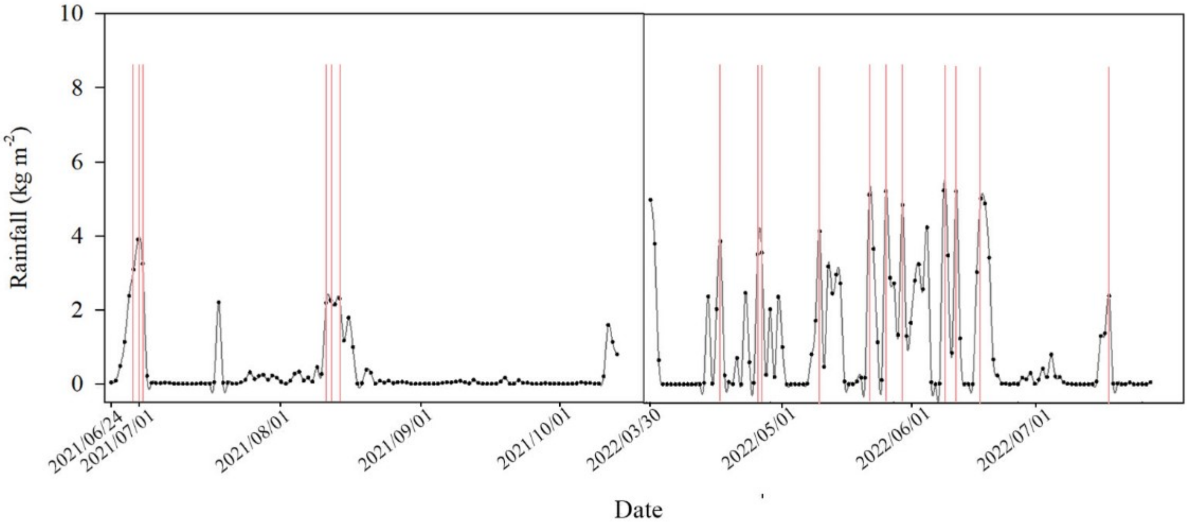

Figure S1. Rainfall data are based on historical re-analysis datasets from the European Centre for Medium-Range Weather Forecasts (ECMWF)/National Aeronautics and Space Administration (NASA), provided by www.xihe-energy.com. The vertical pink line indicates the time when the mesocosms were covered by a temporary roof.

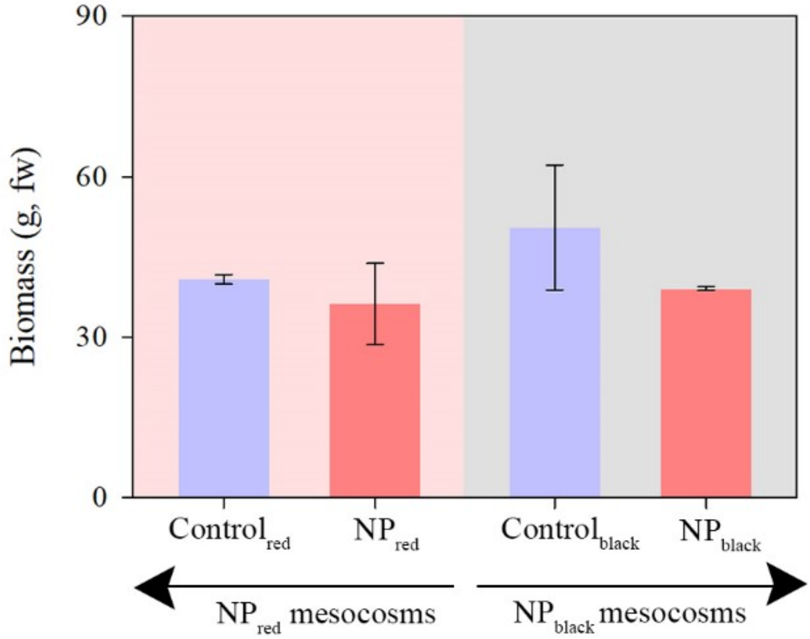

Figure S2. The fresh weight of radish roots from NP$_{red}$ and NP$_{black}$ mesocosms. NP$_{red}$: mesocosms with NPs in red soil; NP$_{black}$: mesocosms with NPs in black soil. Data shown are the averages of two replicates ± SD. All individuals were pooled together for each replicate. No significant difference was observed between the treatments at $p > 0.05$.

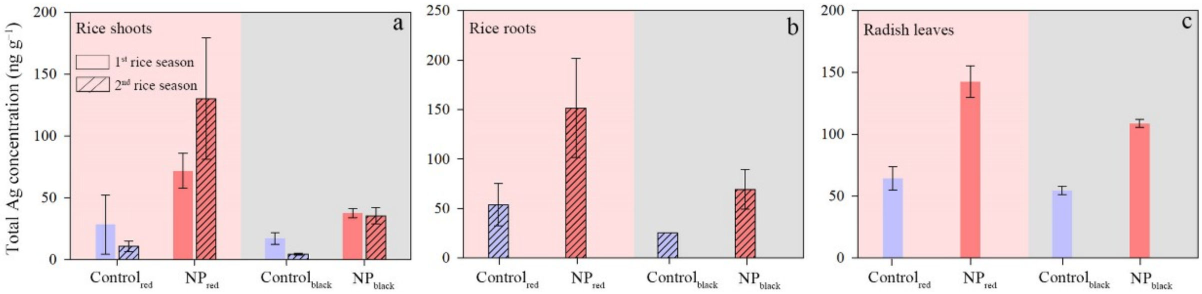

Figure S3. Bioaccumulation of Ag in (a) rice shoots, (b) rice roots, and (c) radish leaves after exposure to NPs. NP_{red}: mesocosms with NPs in red soil; NP_{black}: mesocosms with NPs in black soil. Data shown are the averages of four replicates ± SD for rice shoots and radish leaves, and the averages of two replicates ± SD for rice roots.

Figure S4. The available data on the levels of soil-borne NPs. The data are available in the literature.[1-3]

Table S1. Physical and chemical properties of the sampled soils.

Sample	Texture	Organic matter content (g kg^{-1})[a] *	pH[b] *	CEC[c] (mol kg^{-1})	Total nitrogen[d] (%)	Total Ag (mg kg^{-1})*	Total Fe (g kg^{-1})*
Red soil	Loamy clay	16.9 ± 0.03	5.0 ± 0.04	12.3	0.16	0.4 ± 0.01	37.1 ± 1.7
Black soil	Loamy clay	27.6 ± 0.1	6.7 ± 0.1	29.0	0.15	0.2 ± 0.01	28.6 ± 0.9

[a] Determination of organic matter by potassium dichromate oxidation and external heating. Data shown are the averages of two replicates ± SD (n=2).

[b] pH was determined by potentiometric method with soil-water ratio 1:5. Data shown are the averages of two replicates ± SD (n=2).

[c] Determination of cation exchange capacity (CEC) by EDTA-ammonium salt.

[d] Determination of total nitrogen by semi-trace Kjeldahl method.

*A significant difference was observed between the soils ($p<0.05$).

Table S2. Concentrations of major cations in the water column before NPs addition. The water was collected from a local Reservoir. The cations were measured by Inductively Coupled Plasma Optical Emission Spectrometry (iCAP7400, Thermo Fisher, USA). Data shown are the averages of four replicates ± SD.

Cations (mg L^{-1})	NP_{Red} mesocosm	NP_{Black} mesocosm
K^{+} *	2.5 ± 0.7	0.8± 0.4
Ca^{2+} *	8.3 ± 0.7	76.1 ± 8.2
Mg^{2+} *	5.2 ± 1.0	13.8 ± 1.0

* A significant difference was observed between the soils (*p*<0.05).

Table S3. The isotope abundances of $^{109}Ag_2S$-NPs and Ag_2S-NPs.

Isotope	Abundance	
	Ag_2S-NPs[a]	$^{109}Ag_2S$-NPs[b]
^{107}Ag	0.5180	0.0042
^{109}Ag	0.4820	0.9958

[a] The abundance of ^{107}Ag and ^{109}Ag in Ag_2S-NPs are the natural abundances of Ag, respectively.[4]

[b] The abundances of ^{107}Ag and ^{109}Ag in $^{109}Ag_2S$-NPs are calculated based on the determined concentrations of ^{107}Ag and ^{109}Ag by ICP-MS. Data shown are the averages of ten replicates.

Table S4. The relative bioavailability of water-borne to soil-borne NPs. This is calculated as the percentage of biotic Ag from initially water-borne NPs (i.e., normalized by their exposure concentrations) divided by the percentage of biotic Ag from the soil-borne NPs.

Soil type	Plant	Time	Relative bioavailability of water-borne NPs to soil-borne NPs
NP_{red} mesocosms	Brown rice	1st rice season	2.4 ± 1.5[a]
		2nd rice season	0.2 ± 0.1[a]
	Duckweed	2nd rice season	8.1 ± 0.01[b]
	Rice borer	2nd rice season	1.3 ± 1.8[b]
NP_{black} mesocosms	Brown rice	1st rice season	3.1 ± 0.6[a]
		2nd rice season	1.8 ± 0.6[a]
	Duckweed	2nd rice season	6.8 ± 2.8[b]
	Rice borer	2nd rice season	2.8 ± 1.0[b]
NP_{red} mesocosms	Radish roots	-	2.6 ± 0.5[a]
NP_{black} mesocosms	Radish roots	-	4.8 ± 0.7[a]

[a] Data shown are the averages of four replicates ± SD.

[b] Data shown are the averages of two replicates ± SD.

Table S5. The soil-to-plant transfer factor for soil-borne NPs (TF, the ratio of Ag concentrations in edible tissues derived from soil-borne NPs to their soil-borne NPs levels).

Soil type	Plant	Time	TF value
NP_{red} mesocosms	Brown rice	1st rice season	2.1×10^{-2}
		2nd rice season	1.4×10^{-2}
NP_{black} mesocosms	Brown rice	1st rice season	1.8×10^{-2}
		2nd rice season	4.6×10^{-3}
NP_{red} mesocosms	Radish roots	-	7.4×10^{-2}
NP_{black} mesocosms	Radish roots	-	1.5×10^{-2}

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