## Supporting information (SI)

Nano-activated Carbon Reduces Mercury Mobility and Uptake by Oryza

Sativa L: Mechanistic Investigation Using Spectroscopic and Microscopic

Techniques

Jianxu Wang  $^{*,\dagger,\ddagger,\$}$ , Sabry M. Shaheen  $^{\ddagger,\psi,\P}$ , Christopher W. N. Anderson  $^{\dagger}$ , Ying Xing

<sup> $\zeta$ </sup>, Shirong Liu<sup>†</sup>, Jicheng Xia<sup>†</sup>, Xinbin Feng<sup>\*,†, §</sup>, Jörg Rinklebe <sup>‡,\*</sup>

<sup>†</sup>State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, 550081 Guiyang, P.R. China; E-mail: wangjianxu@vip.gyig.ac.cn (J.X); fengxinbin@vip.skleg.cn (X.B)

<sup>‡</sup>University of Wuppertal, School of Architecture and Civil Engineering, Institute of Foundation Engineering, Water- and Waste-Management, Laboratory of Soil- and Groundwater-Management, Pauluskirchstraße 7, 42285 Wuppertal, Germany; E-mail: <u>shaheen@uni-wuppertal.de</u> (Shaheen); <u>rinklebe@uni-wuppertal.de</u>(Rinklebe) <sup>§</sup>CAS Center for Excellence in Quaternary Science and Global Change, Xi'an, 710061, China.

<sup>\*</sup> King Abdulaziz University, Faculty of Meteorology, Environment, and Arid Land Agriculture, Department of Arid Land Agriculture, Jeddah 21589, Kingdom of Saudi Arabia

<sup>®</sup>University of Kafrelsheikh, Faculty of Agriculture, Department of Soil and Water Sciences, 33516 Kafr El-Sheikh, Egypt; E-mail: <u>smshaheen@agr.kfs.edu.eg</u> <sup>®</sup>Environmental Sciences, School of Agriculture and Environment, Massey University, 4442 Palmerston North, New Zealand; E-mail:

C.W.N.Anderson@massey.ac.nz

<sup>ζ</sup> School of Chemistry and Materials Science, Guizhou Normal University, 550001 Guiyang, China;E-mail:<u>xy31034@163.com</u>

\*University of Sejong, Department of Environment, Energy and Geoinformatics, 98 Gunja-Dong, Guangjin-Gu, Seoul, Republic of Korea

\*Corresponding authors: wangjianxu@vip.gyig.ac.cn; fengxinbin@vip.skleg.cn

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#### S1 Sample analysis

#### S1.1 Soil samples

The pH value of soil was determined using a pH meter (Hanna HI3M, Hanna instruments®, USA) in its suspension with solid to deionized water ratio of 1:2.5 (w/w). The total Hg content in the soil was directly measured by pyrolysis of samples at 600 °C to 700 °C using Lumex RA 915+ coupled with a Pyro 915 pyrolysis attachment, which has a determination limit of 5 ng  $g^{-1}$ . Total carbon, total nitrogen, and total sulphur were directly measured using an Elemental Analyzer (PE2400-II, MA, USA). The total contents of Fe was determined by digesting 0.6 g soil in a microwave system (Milestone MLS 1200 Mega, Sorisole, Italy) with 1 mL concentrated HCl (32%) and 3 mL concentrated HNO<sub>3</sub> (65%); the concentrations of Fe in the digested solutions were determined by inductively-coupled optical emission spectrometry (Ultima 2, Horiba Jobin Yvon, Unterhaching, Germany). As for Fe L-edge XANES spectroscopy analysis, Fe reference compounds, including Goethite, FeO,  $Fe_2O_3$ ,  $Fe_3O_4$ , FeS, FeSO<sub>4</sub>, pyrite,  $Fe(OH)_3$ , Ferrihydrite,  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>, and Lepidocrocite, were analyzed at Beamline 4WB1 at the Beijing Synchrotron Radiation Facility. The soil pellet and Fe reference compounds were mounted on Scotch tape and fixed to a sample holder. All the samples and standards were measured under florescence mode using a Lytle fluorescence ionization chamber. Spectra were collected at a stepwise of 0.3 eV from 650 to 750 KeV. The XANES L-edge spectral of the standards and samples were plotted with the energy ranged

between 680 and 740 eV.

#### S1.2 Nano-activated carbon (NAC)

Total carbon content of the NAC was analyzed using an Elemental Analyzer (PE2400-II, MA, USA). The specific surface area of the particle was determined by the Brunauer–Emmet–Teller (BET) method using an N<sub>2</sub> adsorption/desorption analyzer (Autosorb-iQ2, Quanta-Chrome Instruments, USA). Nano-activated carbon powders were dispersed with 50% ethanol, mounted in a carbon-covered copper grid, and analyzed using an analytical transmission electron microscope (Tecnai G2 F20 S-TWIN TMP, FEI Co., America) operated at 200 kV.

#### S1.3 Liquid samples

Subsamples for total dissolved Hg analysis were acidified with concentrated hydrochloric acid (HCl) (2%, v/v), and the Hg concentration was determined according to USEPA 1631<sup>1</sup>. Sulfate and DOC were analyzed using a ICS-90 Ion Chromatography System (Dionex Co., USA) and a Elementar High TOC II analyzer (Elementar Co., Germany), respectively. The oxidation-reduction potential (ORP) and pH of pore water was measured immediately by an ORP meter (AZ8552, Henxin Co., Taiwan) and a pH meter (Hanna HI3M; Hanna Instruments INC, RI, USA), respectively, prior to sub-sampling (The pore water collected on day 5 was not analyzed for ORP).

#### S1.4 Plant samples

For plant sample total Hg determinations, about 0.1 g plant powders were pyrolyzed at 600°C to 700°C in a PYRO-915 attachment (Lumex, St. Petersburg, Russia), and the volatilized Hg concentration was measured using a Lumex RA 915+. The dry weights of leaf, stalk, and root were measured using a balance with a precision of 0.01g (BSM, Shanghai zhuojing electronic Co., China). The longest lengths of root and stalk were manually measured by a ruler. The volume of fresh roots was measured using the water-displacement method  $^2$ .

# S2 Mercury L<sub>3</sub>-edge and sulfur K-edge XANES spectroscopy

As for Hg L<sub>3</sub>-edge XANES spectroscopy analyses, an energy range of 12.18-12.58 KeV was used to acquire the spectra. Data for Hg reference compounds were collected in transmission mode (Hg concentration>5%), and for sample pellets in fluorescence mode using a 19-element high-purity Ge solid-state detector under ambient conditions (soil matrix; 50 mg kg<sup>-1</sup><Hg concentration<0.1%). Data normalization (background correction), and linear combination fitting (LCF) was performed with the IFEFFIT software package <sup>3</sup>. The Hg L<sub>3</sub>-edge XANES spectra of the standards and samples were plotted with the energy ranged between 12.20 and 12.58 KeV.

As for S K-edge XANES spectroscopy analyses, all standards and samples were measured under florescence mode in total electron yield mode using a fluorescent ion chamber Si (Li)detector (PGT LS30135). The K-edge of FeSO<sub>4</sub> positioned at 2482.5 eV was used for energy calibration. Spectra were collected at a stepwise interval of 0.3 eV from 2.25 to 2.6 KeV. The XANES K-edge spectra of the standards and samples were plotted with an energy range between 2.46 and 2.5 KeV.

# S3 Cost-effectiveness analysis of using nano-activated carbon for Hg-contaminated farmland remediation

Assuming that the target soil depth for remediation is 20 cm (About 90% of biomass of roots distributes at this soil layer <sup>4</sup>), the soil density might be 1.0 g cm<sup>-3 5</sup>, the application rate of amendment is 1 kg nano-activated carbon per 100 kg of soil (1%), and the treating of 1 hectare of farmland (10, 000 m<sup>2</sup>) needs about 20 ton of nano-activated carbon. The price of nano-activated carbon is about 880 US\$/ton (the price was available at Alibaba.com). The rough capital cost for remediation of Hg-contaminated farmland is 17,600 US\$/ hectare. According to a Chinese report, the current techniques used for heavy metal-contaminated farmland remediation consume about 40,000 US\$/ hectare (available at https://www.globalelr.com/2018/08/china-announces-control-standard-for-soil-conta mination/). Thus, the capital cost for using nano-activated carbon in Hg-contaminated farmlands remediation might be acceptable.

#### **Supplementary Figures**



Fig S1 The Fe L-edge XANES spectra of the studied soil and  $\alpha\text{-}\text{Fe}_2\text{O}_3$ 



Fig S2 Transmission electron microscopy (TEM) image of the studied nano activated carbon (NAC) aggregates.



Fig S3 The length of root and aboveground tissue, and the volume of root; differences at P<0.05 tested by least significant difference in one-way analysis of variance in each biomass parameter between control and nano-activated carbon (NAC) treatments are

indicated by different small-case letters on each bar.



Fig S4 The concentration of sulfate in the pore water in control and nano activated carbon (NAC) treatments throughout the rice growing season.



Fig S5 The oxidation reduction potential of the pore water in control and nano activated carbon (NAC) treatments throughout the rice growing season.



Fig S6 the relationship between oxidation reduction potential (ORP) and sulfate in the pore water throughout the rice growing season; The sulfate concentration in the pore water collected on day 5 was not included in the analysis due to the lack of ORP data.



Fig S7 The pH value of pore water in control and nano-activated carbon (NAC) treatments throughout the rice growing season.



Fig S8 The comparison of total Hg content in soil (A), total dissolved Hg (THg) in pore water (B), soil: water partition coefficients ( $K_d$ )(C), Hg contents in plant tissues (D), BAF (E), and Hg mass in plant tissues (Hg<sub>root+stalk+leaf</sub> mass) (F), between control and nano-activated carbon (NAC) treatments. Error Bars denote standard deviation

from means ofthree replicates (1SD); significant differences among control and nano-activated carbon treatments are indicated by asterisks (P<0.05), evaluated based on ANOVA and Least Significant Difference (LSD) comparisons.



Fig S9 The relationship between dissolved Hg and dissolved organic carbon (DOC) concentration in the pore water in all treatments throughout rice growing season.



Fig S10 A: the relationship between Hg contents in tissues of rice plant and Hg concentration in the pore water; B: the relationship between  $K_d$  and BAF. Data for plant tissue Hg content is obtained by the summarization of Hg contents in different tissues of rice plant (Root, stalk, and leaf); Data for pore water Hg concentration is obtained by averaging its Hg concentrations throughout rice growing season; please

note that the data for  $K_d$  and BAF were transformed using Ln functions.



Fig S11 EDX spectra for the soil micro-aggregates as indicated in Fig 4, A, B, C



Fig S12 Identification of Hg-S clusters in 3% nano-activated carbon treatment using Transmission electron microscopy coupled with energy dispersive X-ray (TEM-EDX) spectroscopy. The EDX spectra figures ①,②,③, and ④ were collected from particles showed in TEM images marked with ①,②,③, and ④. Particles circled by dashed lines with blue colour are nano-activated carbons (nC), with dark green colour are chlorites (cH), with yellow colour is organic matter (oM), and with red colour are Hg-S aggregates.

# **Supplementary Tables**

the studied soli. The $n-3$				
		Soil		
	7.5±0.2			
Total	20±1.3			
Total N (mg g <sup>-1</sup> )		2.1±0.1		
Total S (mg g <sup>-1</sup> )		1.0±0.2		
Particle	Clay ( $< 2 \mu m$ )	3.4%		
Size	Silt (2 $\sim$ 50 $\mu$ m)	32.3%		
distribution	Sand ( $> 50 \ \mu m$ )	54.3%		
Total Hg (mg kg <sup>-1</sup> )		129±24		

Table S1 The selected physico-chemical properties of

the studied soil mean+sd (n=3)

Table S2 The selected physico-chemical properties of the studied nano-activated carbon (NAC) (n=3)

Properties	Diameters	Specific surface	Total C	Density
	(nm)	area $(m^2 g^{-1})$	(%)	$(g \text{ cm}^{-3})$
NAC	20-50	500	99.5	3.02

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