Supplementary Information

Deciphering and Predicting Microscale Controls on Radon Production in Soils, Sediments and Rock

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Sequential Extractions:

S1: 1.0 gram composite shale sample was shaken with 16 ml of 1 M $MgCl_2$ (pH 7) on a rotary shaker for 1 h at room temperature. Thereafter, sample was centrifuged at 6000 rpm for 15 min and supernatant was filtered. The solid residue was rinsed with 10 ml of de-ionized water (18 MΩ), which after centrifugation was collected and added to the reagent solution to be analyzed.

S2: The residue was shaken with 25 ml of 1M CH_3COONa (pH 5) for 5 h at room temperature, centrifuged, and the supernatant filtered. Residue was rinsed with de-ionized water as in step S1 and added to reagent solution to be analyzed.

S3: To the residue, 20 ml of 0.04 M $NH_2OH.HCL$ in 25% w/w CH_3COOH is added, whereafter the suspension shaken for 6 h at 96 ^oC, centrifuged, and supernatant filtered. Residue was rinsed with de-ionized water as in step S1 and added to reagent solution to be analyzed.

S4: To the residue, 3 ml of 0.02 M HNO_3 and 5 ml H_2O_2 (30% w/v) was added and allowed to react for 2h at 85 ^oC. A second aliquot of 3 ml of H2O2 (30% w/v) was added and allowed to react for 3 h at 85 ^oC. After cooling the solution to room temperature, 5 ml of 3.2 M of CH_3COONH_4 was added to solution and allowed to react for 30 min and filtered.

S5: The shale residue after the oxidizable extraction step (S4) primarily contains silicate minerals; residue composition was estimated from the difference between the whole-rock digestions and the sum of the sequential fractions (S1-S4).

1. **Steady state calculation of Radon Production rate**

Assumptions:

1) The volumetric distribution of ^{226}Ra is uniform within the solid
2) Large scale diffusion and advection are neglected for this analysis
3) Secular Equilibrium conditions are assumed, i.e.

$$^{226}_{Ra}A = {}^{222}_{Rn}A$$

Notation:

A= Specific activity of ^{226}Ra, (Bq m^{-3} of solid volume)

S= surface area of solid, $m^2\,g^{-1}$

M= Mass of the solid, grams

R_1=recoil range of ^{222}Rn, meters

Z= depth from the surface of the grain, meter

ε_1 = probability of escape of recoiled ^{222}Rn entering pore space, given by $= \frac{(R_1 - Z)}{2R_1}$ (Fleischer, 1983; Semkow, 1989)

The rate of production of ^{222}Rn **P** from direct recoil from ^{226}Ra in solid can be given by:

$$\mathbf{P} = \frac{\left(\int_0^{R_1} \mathrm{ASM}\, \varepsilon_1 \mathrm{d}Z\right)}{V_p} \qquad \mathrm{Bq\ m^{-3}}$$

$$= \frac{\int_0^{R_1} \mathrm{ASM} \frac{(R_1 - Z)}{2R_1} \mathrm{d}Z}{V_p}$$

$$= \frac{\mathrm{ASM}}{4V_p R_1}[-0 + R_1^2]$$

$$\mathbf{P} = \frac{\mathbf{ASMR_1}}{\mathbf{4V_p}} \qquad \mathbf{Bq\ m^{-3}\ of\ pore\ space}$$

	Quartz	Microcline	Geothite	Thorite
Stoichiometric formula	SiO_2	$KAlSi_3O_8$	FeO(OH)	$ThSiO_4$
Recoil energy of Rn decay event (keV)	86.2	86.2	86.2	86.2
Compound correction	1	1	1	1
Pure solid density (g cc^{-1})	2.34	2.56	3.65	5.35

Table S1: Input parameters to SRIM code to calculate recoil range of Rn in different minerals. The densities are here of pure mineral and not bulk densities. Atomic number of Si=14, O=8, K=19, Al=13, Fe=26, Th=90 and Rn=86.

U	Exchangeable	Acid Soluble	Reductive	Oxidizable	Residue
Shale (ppm)	6±0.05	ND	5.0±0.5	7.0±1.0	1.0±0.02
Pegmatite (ppm)	139±5	4035±66	ND	3049±37	10974±241

Table S2: Extracted U during each sequential extraction step. The concentrations are reported as average of triplicates and errors denote the standard deviation.

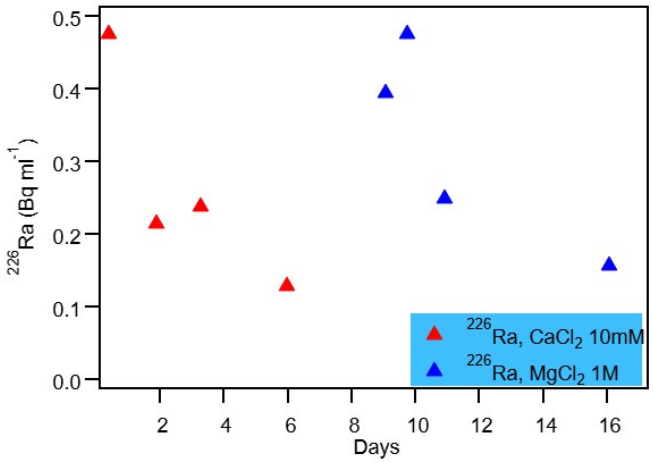

Figure S1: Radium activity in pre-washing stage in granite.

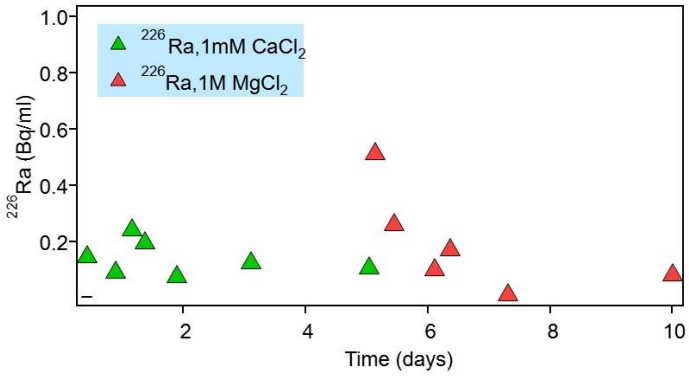

Figure S2: Ra activity in pore fluid during pre-conditioning step in shale.

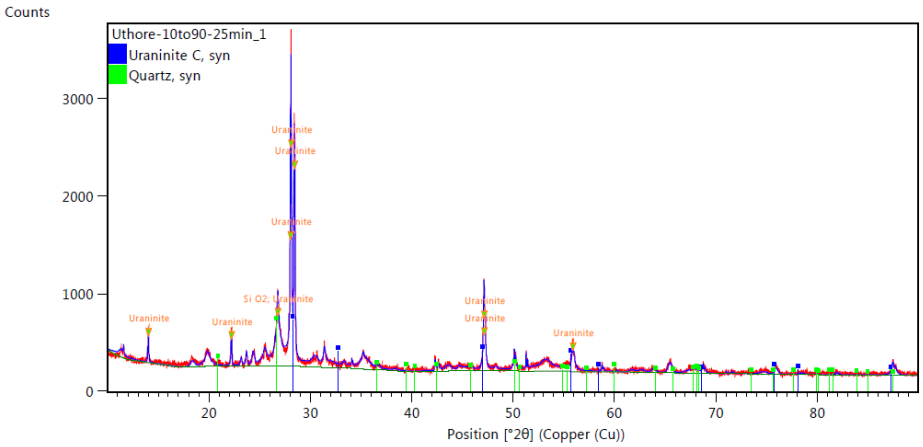

Figure S3: XRD pattern of pegmatite.

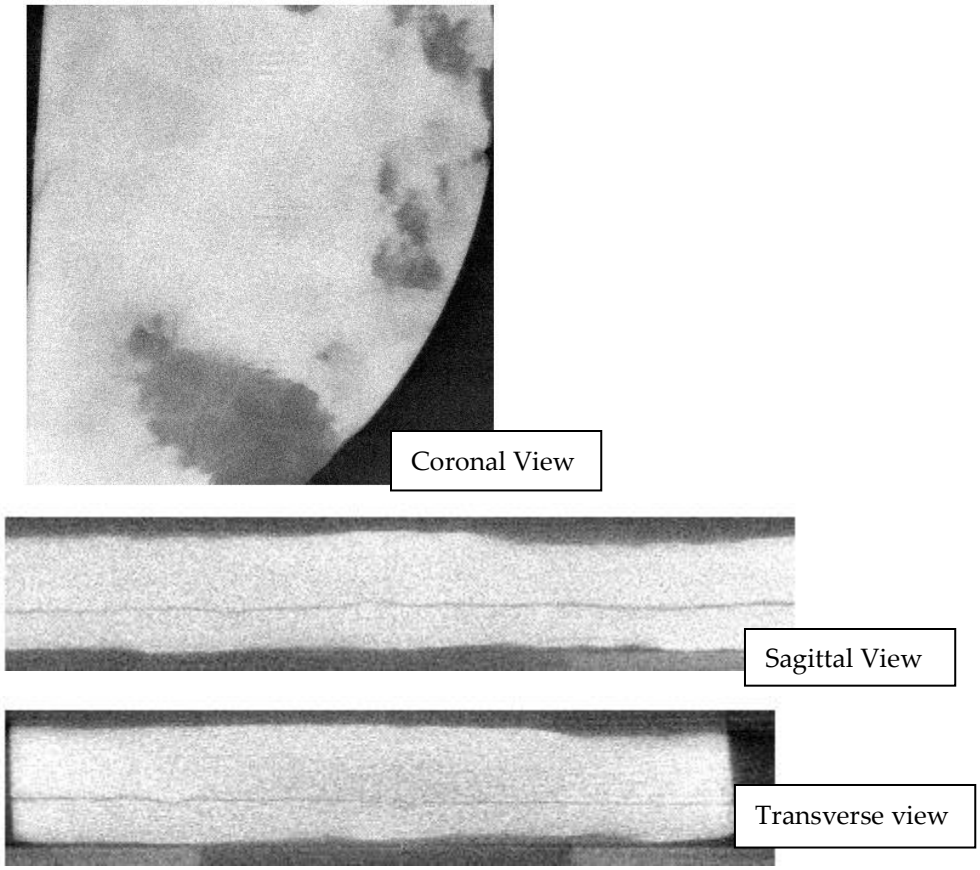

Figure S4: Micro-CT X-Ray imaging of shale. A chipped shale rock was used for the scanning.

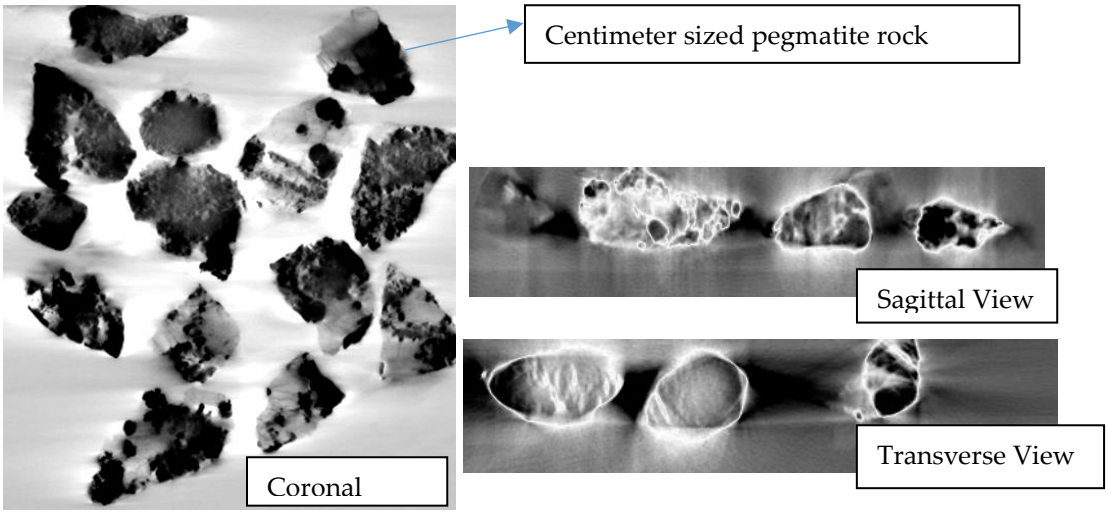

Figure S5: Micro-CT X-Ray imaging of pegmatite.

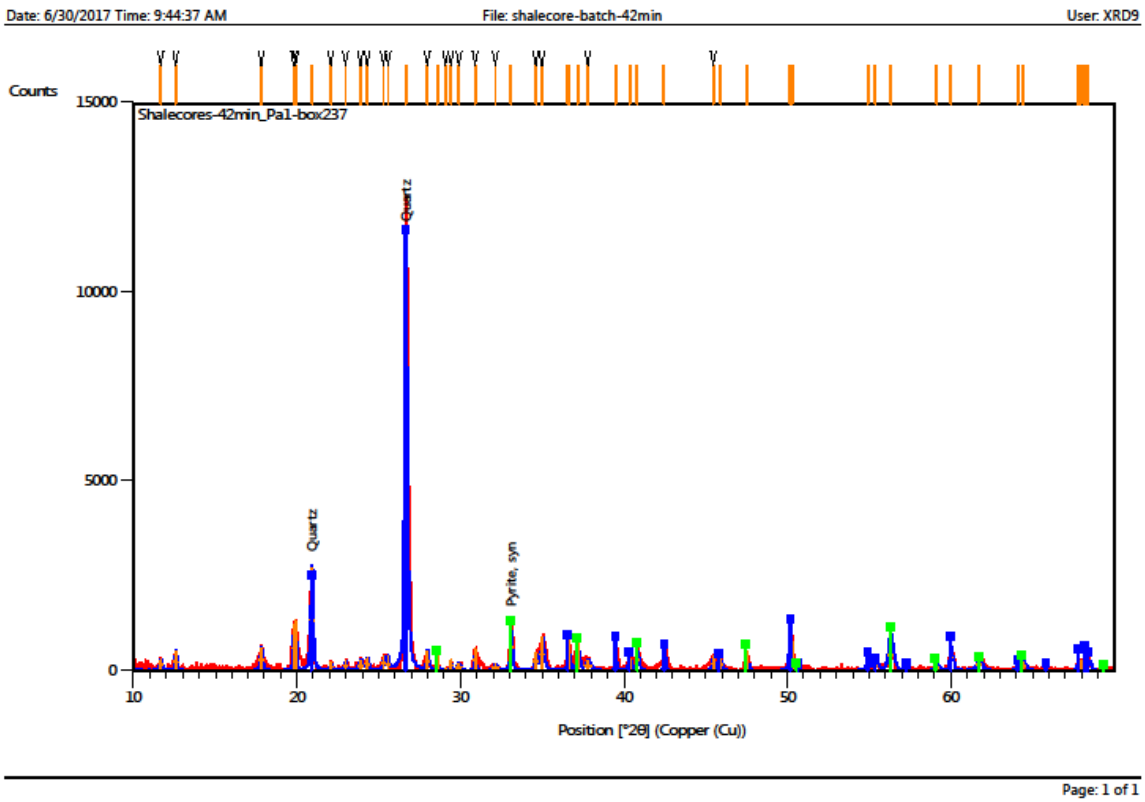

Figure S6: XRD pattern of shale.