Determination of potentially bioavailable Pu in Japanese rice paddy soils with SF-ICPMS

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Outline

- Background
- Method
- Results and discussion
- Perspectives

Background

Soil-rice transfer

- **Soil-rice transfer of Pu: potential radiological impact**
  - FDNPP accident highlighted food safety with radionuclides
  - Rice is the staple food in Japan

- **Transfer factor: estimate bioavailability quantitatively**
  - Traditional transfer factor (TF) for radionuclides

$$TF = \frac{A_P}{A_s} \times 100\%$$

A_p (Bq/kg) and A_s (Bq/kg) are radionuclide activity concentrations in the dry sample of plant tissue and in the soil

  - Available transfer factor (ATF, Beaza et al., 2005):

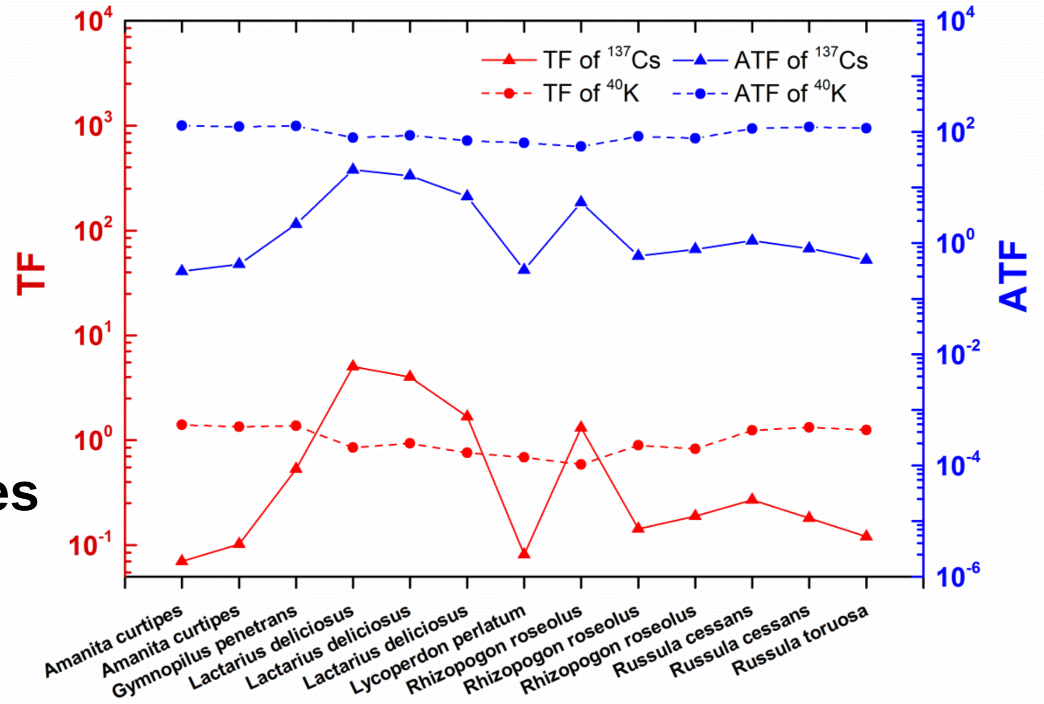
$$ATF = \frac{A_P}{A_s \times AF} \times 100\%$$

AF is the ***bioavailable fraction (%)*** of radionuclide

Only a small fraction of the radionuclide is available for plant uptake!

Considerations between ATF and TF

- **ATF vs. TF for a radionuclide**
  - ATF: considers the part of radionuclide that is “actually” involved in transfer process
  - TF: is it appropriate to apply an averaged TF from literature to a localized case?



Comparison of TF and ATF of soil-fungi transfer for ^{137}Cs and ^{40}K (data from Baeza et al., 2005)

- **ATF vs. TF among radionuclides**
  - ATF reasonably reflects the difference of bioavailability between radionuclides

Main goal of this study:

Investigate the bioavailability of Pu in Japanese rice paddy soils with potentially bioavailable fraction and ATF

Method

Reagents for evaluation of bioavailable Pu

- **Bioavailable fraction of elements in soils**

Bioavailable fraction
- **Water soluble**
  - **H_2O**
- **Exchangeable**
  - **Acid (e.g. HCl, HNO_3)**
  - **Complexing reagents (e.g. EDTA, DTPA)**
  - **Salt solution (e.g. NH_4OAc, $CaCl_2$)**

	Grain				Stalk			
Extraction solution	Cd	Cu	Pb	Zn	Cd	Cu	Pb	Zn
Total	0.326*	0.212	0.437**	0.3535*	0.284*	0.306*	0.374**	0.364**
Mehlich I	0.376**	0.352*	0.367**	0.463**	0.396**	0.476**	0.482**	0.571**
Mehlich III	0.428**	0.296*	0.472**	0.421**	0.406**	0.561**	0.413**	0.506**
EDTA	0.363**	0.416**	0.515**	0.391**	0.426**	0.442**	0.423**	0.565**
DTPA–TEA	0.618**	0.532**	0.466**	0.616**	0.586**	0.496**	0.409**	0.712**
NH_4OAc	0.848**	0.744**	0.834**	0.957**	0.935**	0.930**	0.922**	0.948**
$CaCl_2$	0.832**	0.699**	0.796**	0.867**	0.806**	0.887**	0.892**	0.853**
Soil properties								
Organic carbon	−0.074	−0.033	−0.098	−0.050	0.043	−0.032	−0.021	−0.040
pH	−0.732**	−0.735**	−0.608**	−0.699**	−0.641**	−0.732**	−0.640**	−0.684**
Clay	−0.167	−0.165	−0.071	−0.197	−0.066	−0.178	−0.054	−0.131

Note. * and ** indicate the statistical significance at probability levels of $p < 0.05$ and $p < 0.01$, respectively.

Linear correlation coefficients between heavy metals in rice and extracted fractions (from Zhang et al., 2010)

Neutral salt solution:
better reagent for bioavailability evaluation

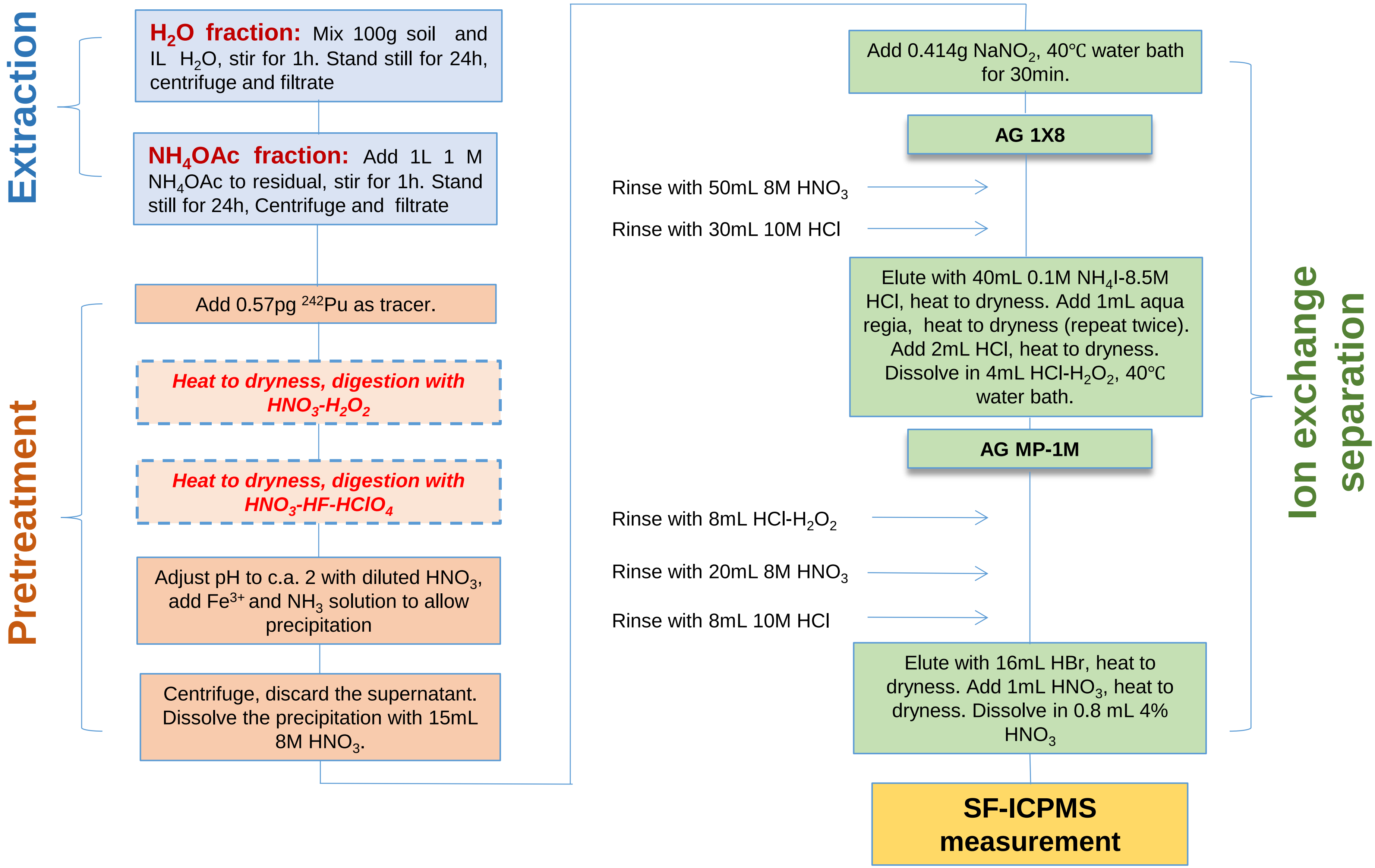
Reagents for evaluation of bioavailable Pu

- **NH_4OAc: widely used to extract exchangeable Pu in soil**

Source	Fraction	Reagent	Radionuclides	Ref.
Artificial solution	exchangeable	1M NH_4OAc	^{239}Pu	Wilson 1966
Artificial solution	exchangeable	1M $MgCl_2$	^{239}Pu	Lee 2002
Chernobyl	water soluble, reversibly bound	H_2O, 1M NH_4OAc	^{238}Pu, $^{239+240}Pu$, ^{241}Am	Ovsiannikova 2010
Chernobyl	exchangeable	1M NH_4OAc	$^{239+240}Pu$, ^{241}Am	Sokolik 2004
Global fallout	exchangeable, diluted acid soluble	1M NH_4OAc, 1M HCl	$^{239+240}Pu$, ^{90}Sr, ^{137}Cs	Guillén 2015
Global fallout	exchangeable, diluted acid soluble	1M NH_4OAc, 1M HCl	$^{239+240}Pu$, ^{90}Sr, ^{241}Am	Baeza 2006
Global fallout/Palomares accident	exchangeable	0.4M $MgCl_2$	$^{239+240}Pu$, ^{90}Sr, ^{137}Cs	Baeza 2005

H_2O and 1M NH_4OAc were employed for the extraction of bioavailable fraction of Pu in soil

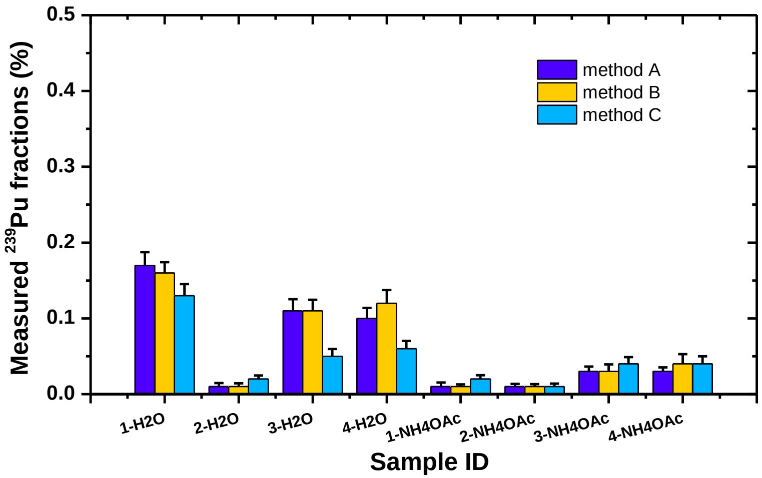
Schematic diagram of the determination of bioavailable Pu



Results and discussion

Comparison of pretreatment methods

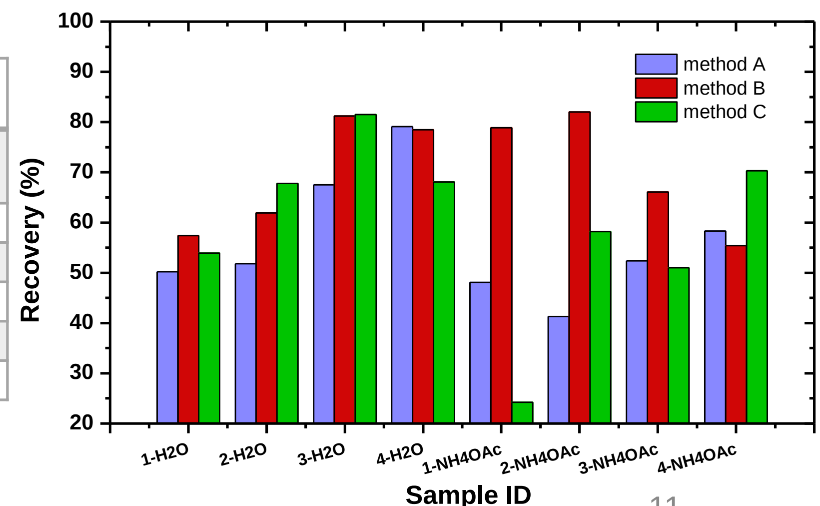
- **Measured ^{239}Pu values were consistent**



- **Method A**: $Fe(OH)_3$ co-precipitation
- **Method B**: acid digestion + $Fe(OH)_3$ co-precipitation
- **Method C**: total digestion + $Fe(OH)_3$ co-precipitation

- **Method B showed the highest recovery**

	Water soluble fraction (%)			Exchangeable fraction (%)		
Sample ID	Method A	Method B	Method C	Method A	Method B	Method C
1	50	57	54	48	79	24
2	52	62	68	41	82	58
3	68	81	82	52	66	51
4	79	79	68	58	55	70
Mean	62±14	**70±12**	68±11	50±7	**71±12**	51±20



Potentially bioavailable Pu in Japanese soils

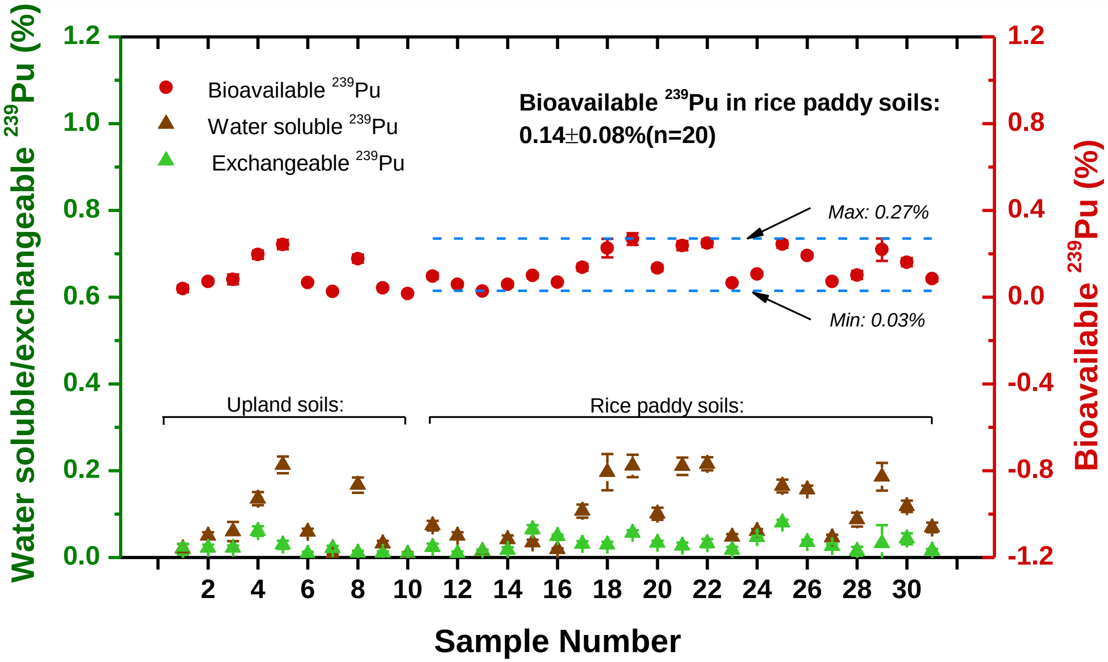
- **Potentially bioavailable Pu in Japanese rice paddy soils:**

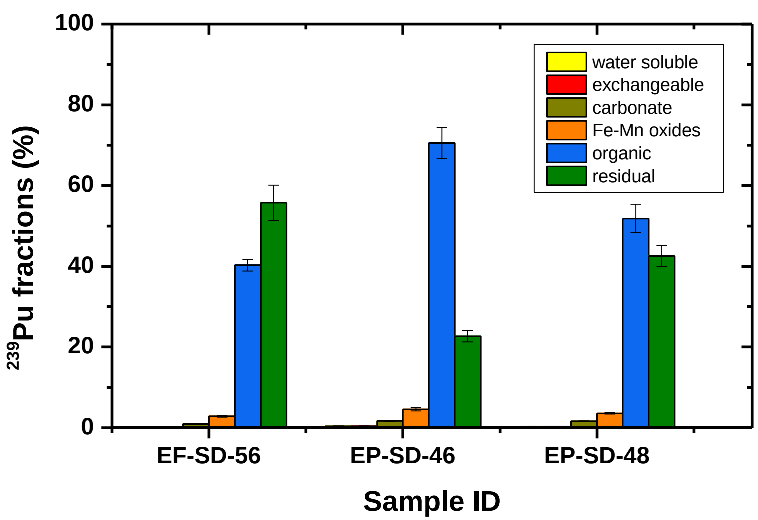
  0.14±0.08% (n=20)

- **No difference was found between upland soils and rice paddy soils**

- **Sequential extraction (modified from Bunzl et al., 1998 and Hou et al., 2003):**

  >90% of Pu associated with organic matter and residual





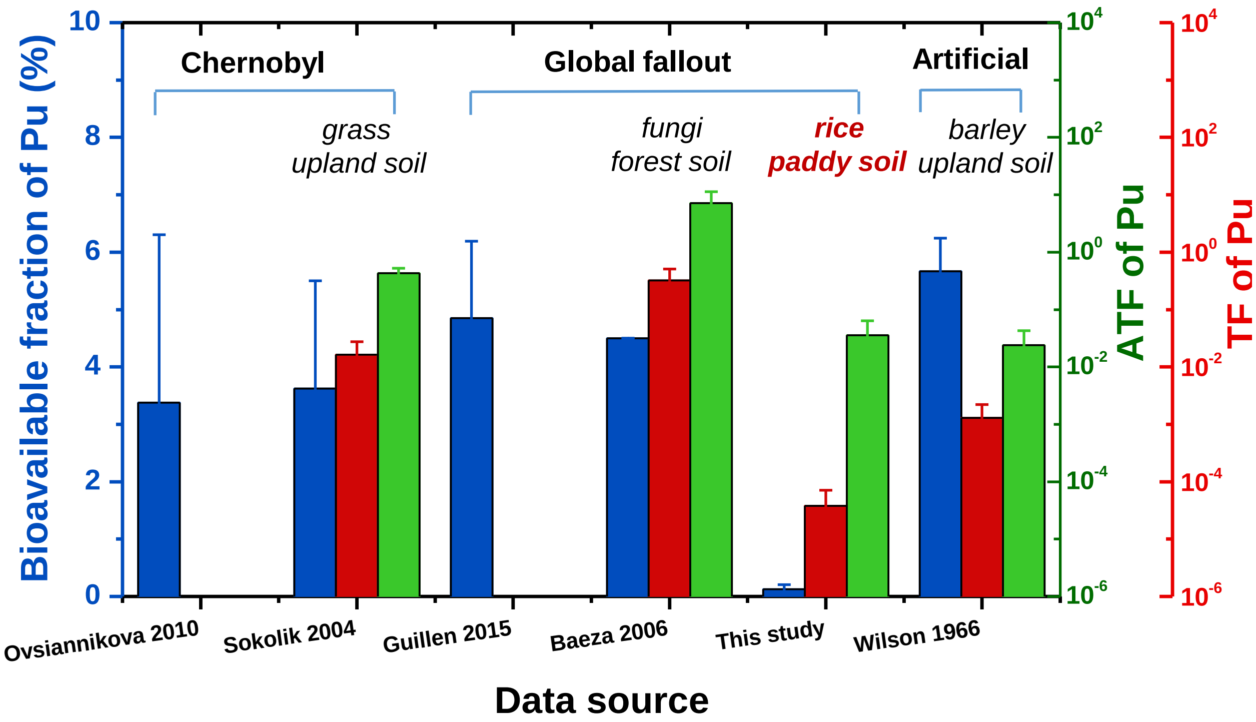
ATF/TF and bioavailable fraction of Pu among ecosystems

- **Bioavailable fraction of Pu varies greatly with ecosystems**

Possible factors

- Source of Pu
- Soil parameters

- **ATF vary with plants**
  - Different plant uptake ability



ATF is a reasonable parameter for evaluation of bioavailability

Thank you