Fukushima's (radio)chemical challenges

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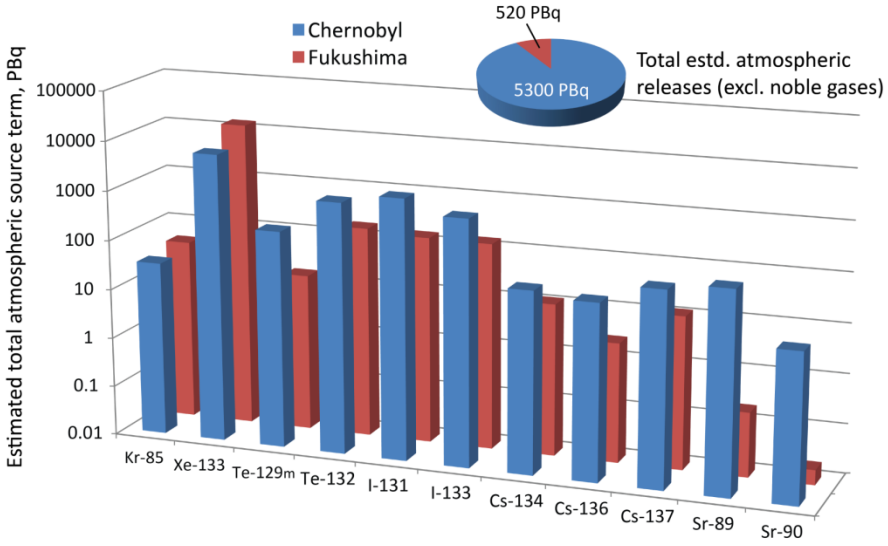
Releases from Fukushima Daiichi NPP

- Total atmospheric release of 520 PBq (excl. noble gases) [1]
- Release of radionuclides of volatile elements Kr, Xe, I, Cs, Te [1]
- Release of less volatile radionuclides (Sr, Pu & actinides): ≪ 1% of total amount
- Volatile radionuclides in environment are well-monitored and documented [2,3]
- Less volatile elements are severely understudied [4]



[1]

[1] G. Steinhauser et al., Sci. Tot. Environ. 470-471 (2014) 800
[2] P. Thakur et al., Sci. Tot. Environ. 458-460 (2013) 577
[3] O. Masson, ..., G. Steinhauser et al., Env. Sci. Technol. 45 (2011) 7670
[4] G. Steinhauser, Env. Sci. Technol. 48 (2014) 4649

Radiocesium signature

- Japanese government ordered analysis of tens of thousands of food samples [1]
- Can we use use this huge data set for environmental research purposes? 445 samples that exceeded regulatory limit [2]
  - Can we find signature of Unit 4 in the environment?
- Fukushima's $^{134}Cs/^{137}Cs$ mean acivity ratio: 0.98 [2]
- Unit 4 was out of operation and defueled since 29 Nov. 2010
- Expected $^{134}Cs/^{137}Cs$ signature approx. 10% lower than 0.98

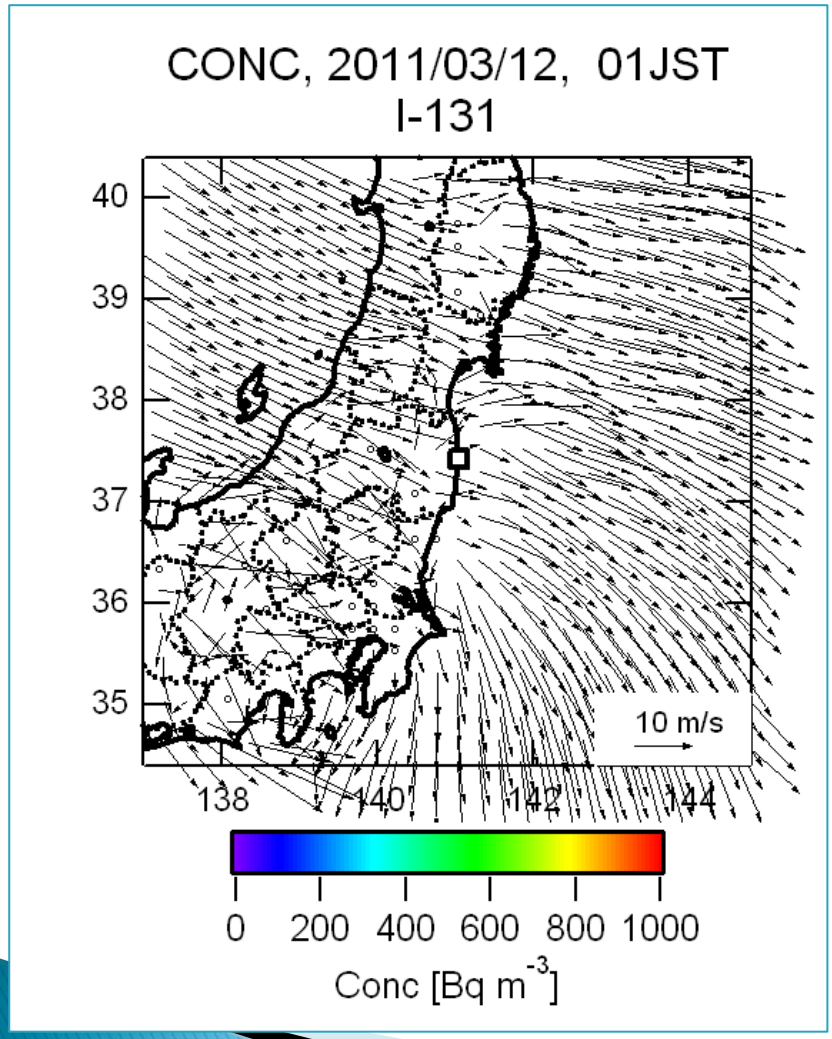
Half-lives
^{134}Cs: 2.06 a
^{137}Cs: 30.17 a

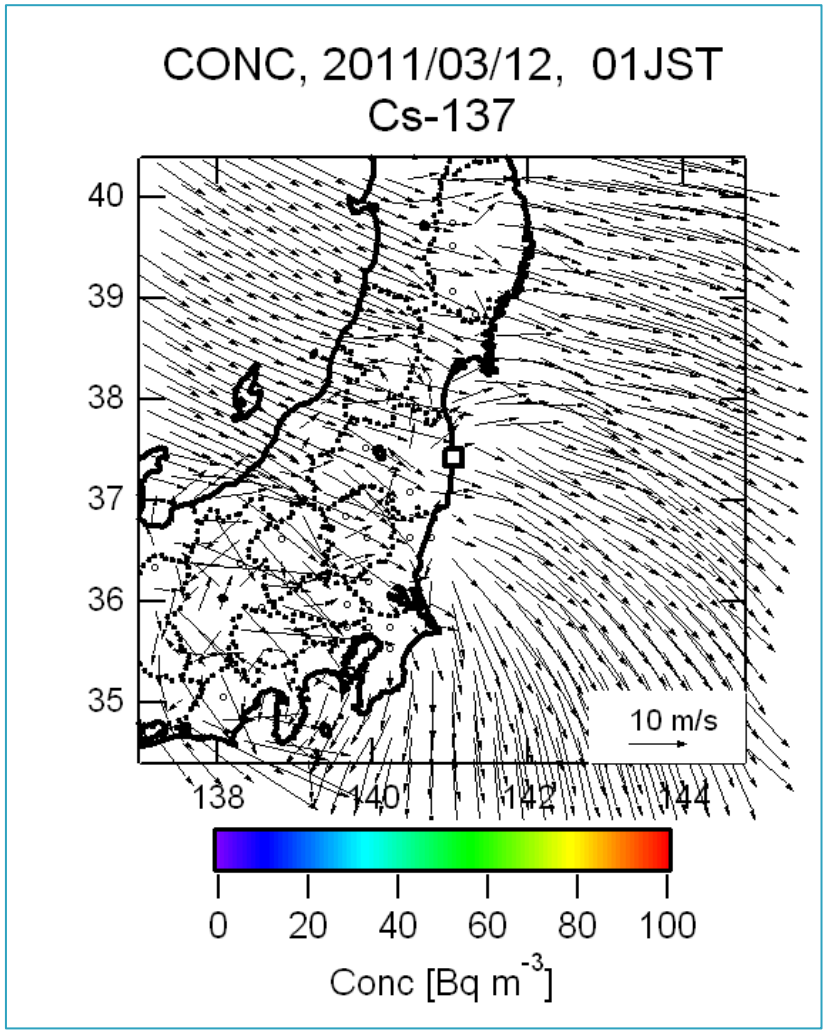
[1] N. Hamada, H. Ogino, J. Environ. Radioactiv. 111 (2012) 83
[2] S. Merz, G. Steinhauser, N. Hamada, Env. Sci. Technol. 47 (2013) 1248

Hydrogen explosions

- Reactor Unit 1: 12 March, 15:31
- Reactor Unit 2: no explosion
- Reactor Unit 3: 14 March, 11:15
- Reactor Unit 4 (**not operational**): 15 March, 6:00

^{131}I and ^{137}Cs atmospheric releases

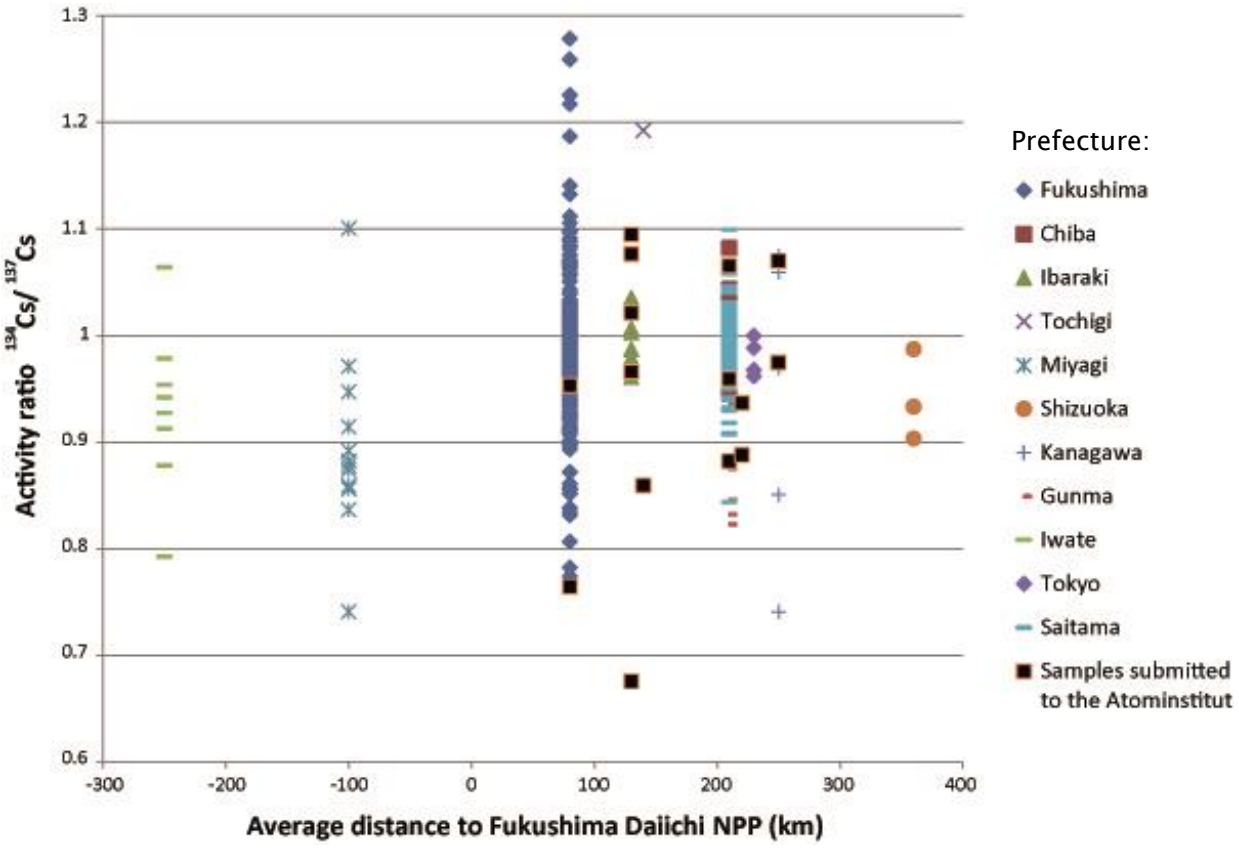




National Institute of Environment, Japan 25 Aug. 2011

Search for Unit 4 signature

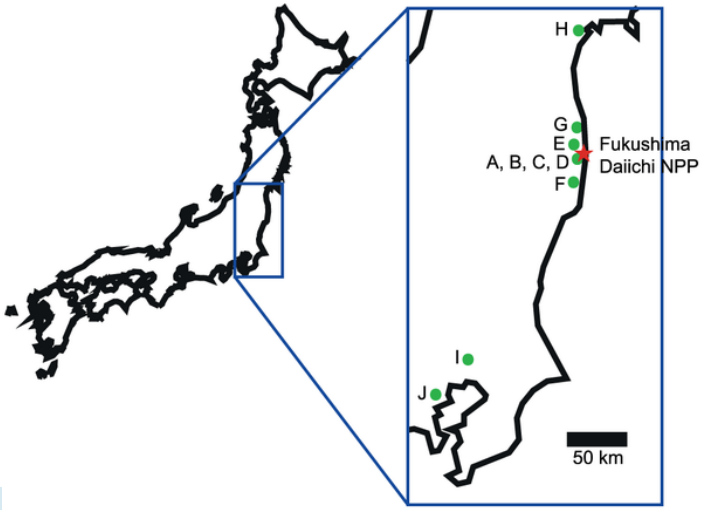
- Decay corrected (to 11-3-11) activity ratio vs. distance to NPP



S. Merz, G. Steinhauser, N. Hamada,
Env. Sci. Technol. 47 (2013) 1248

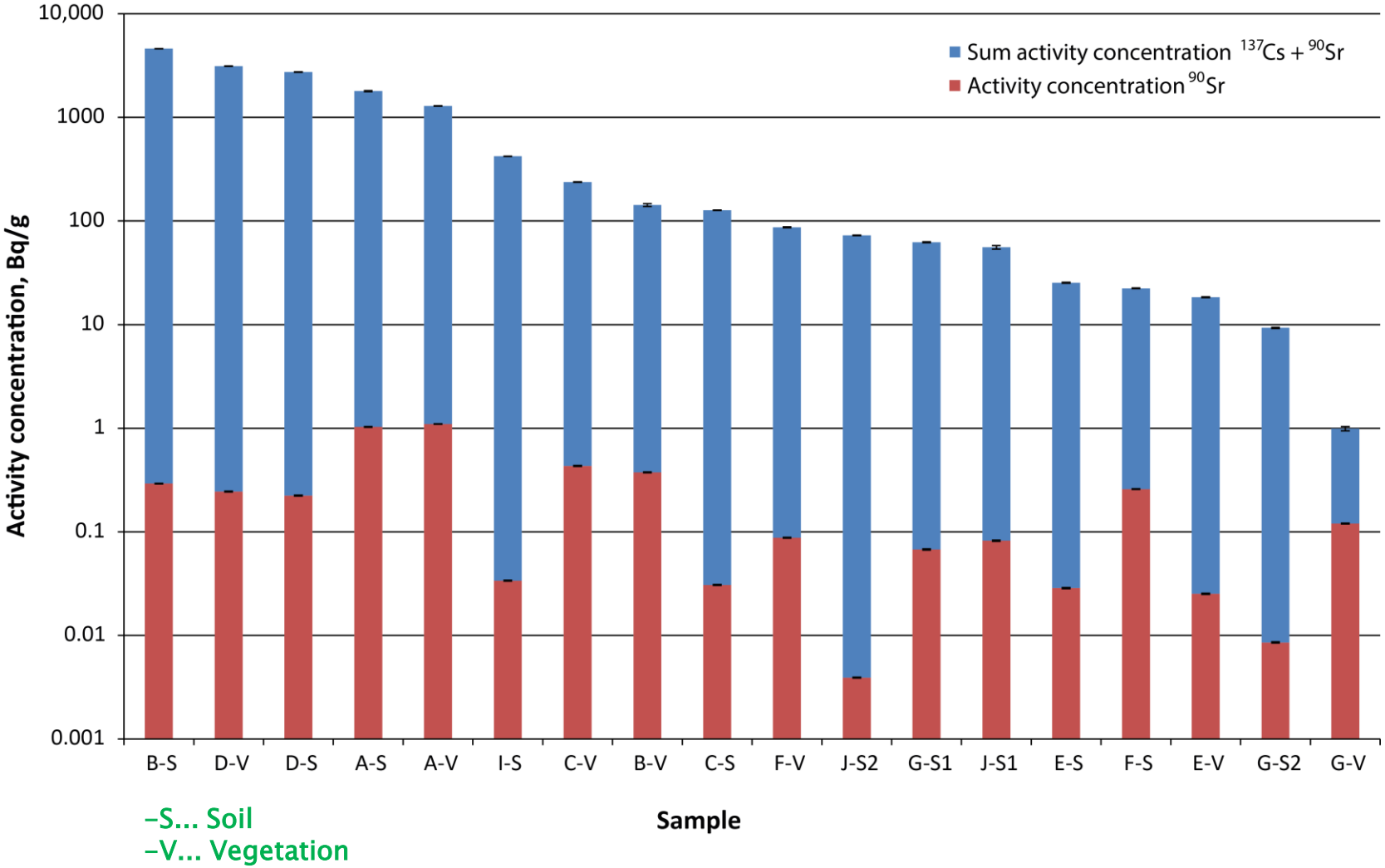
Strontium-90 in the environment

- ^{90}Sr: $T_{1/2}$ = 28.9 y
- Understudied radionuclide
- Intermediate volatility
- Bone-seeker, long-biological half-life, highly dose relevant
- Pure β^- emitter, requires radiochemical separation from coexisting radionuclides



[1] G. Steinhauser et al., PLoS One 8 (2013) e57760

^{90}Sr and ^{137}Cs at selected hot spots (2011)

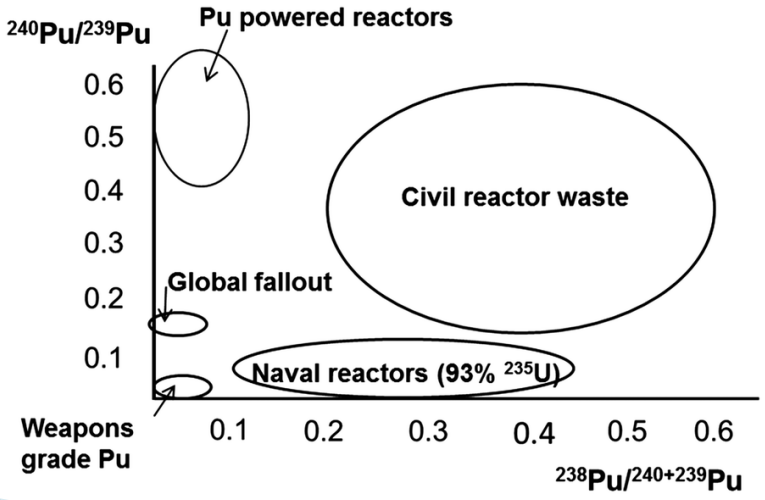


–S... Soil
–V... Vegetation

G. Steinhauser et al., PLoS One 8 (2013) e57760

Plutonium

- We found Pu at many spots inside the exclusion zone [1]
- However, most of it was fallout Pu from nuclear explosions
- Weapon fallout Pu: $^{240}Pu/^{239}Pu$ isotopic ratio 0.18–0.20
- Reactor Pu: $^{240}Pu/^{239}Pu$ isotopic ratio > 0.20

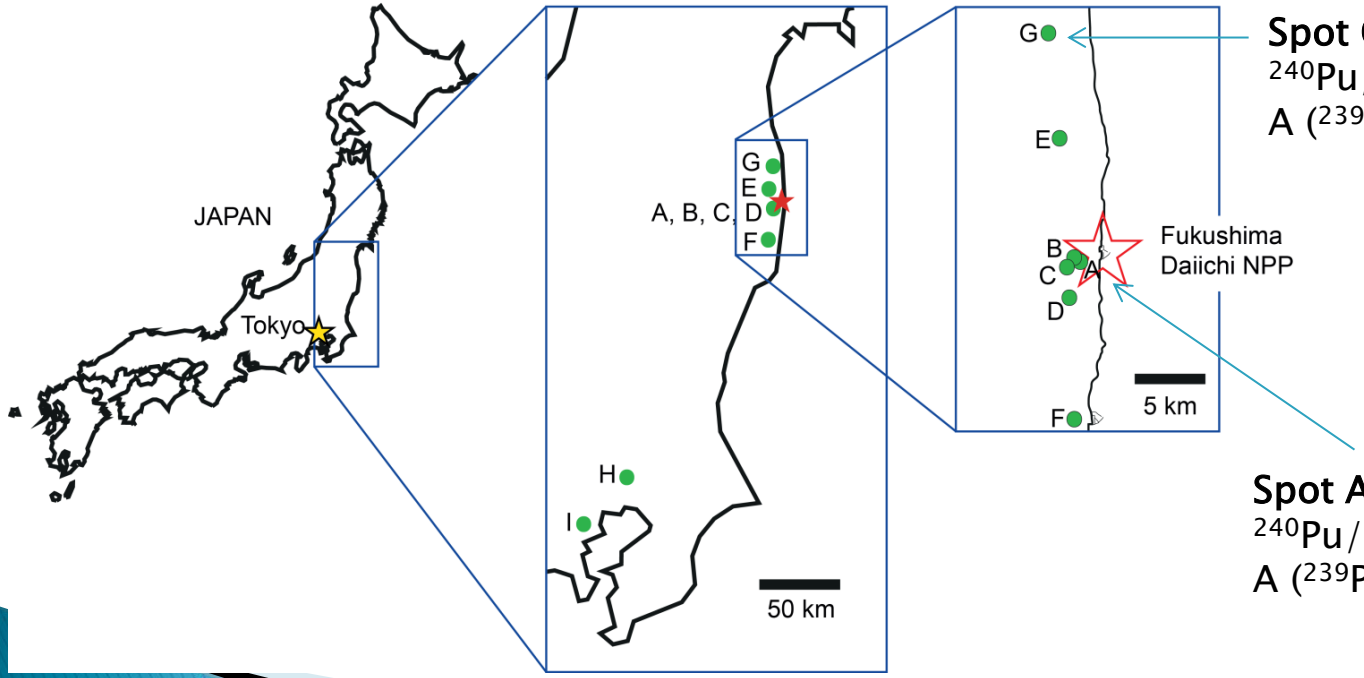


Graph taken from:
S. Cagno et al.,
Env. Sci. Proc. Imp. 16 (2014) 306

[1] S. Schneider,..., G. Steinhauser, Sci. Rep. 3 (2013) 2988

Plutonium

- Radiochemical isolation of Pu (TEVA resin)
- AMS measurement
- Reactor-Pu found in 2 vegetation samples: spot A and G

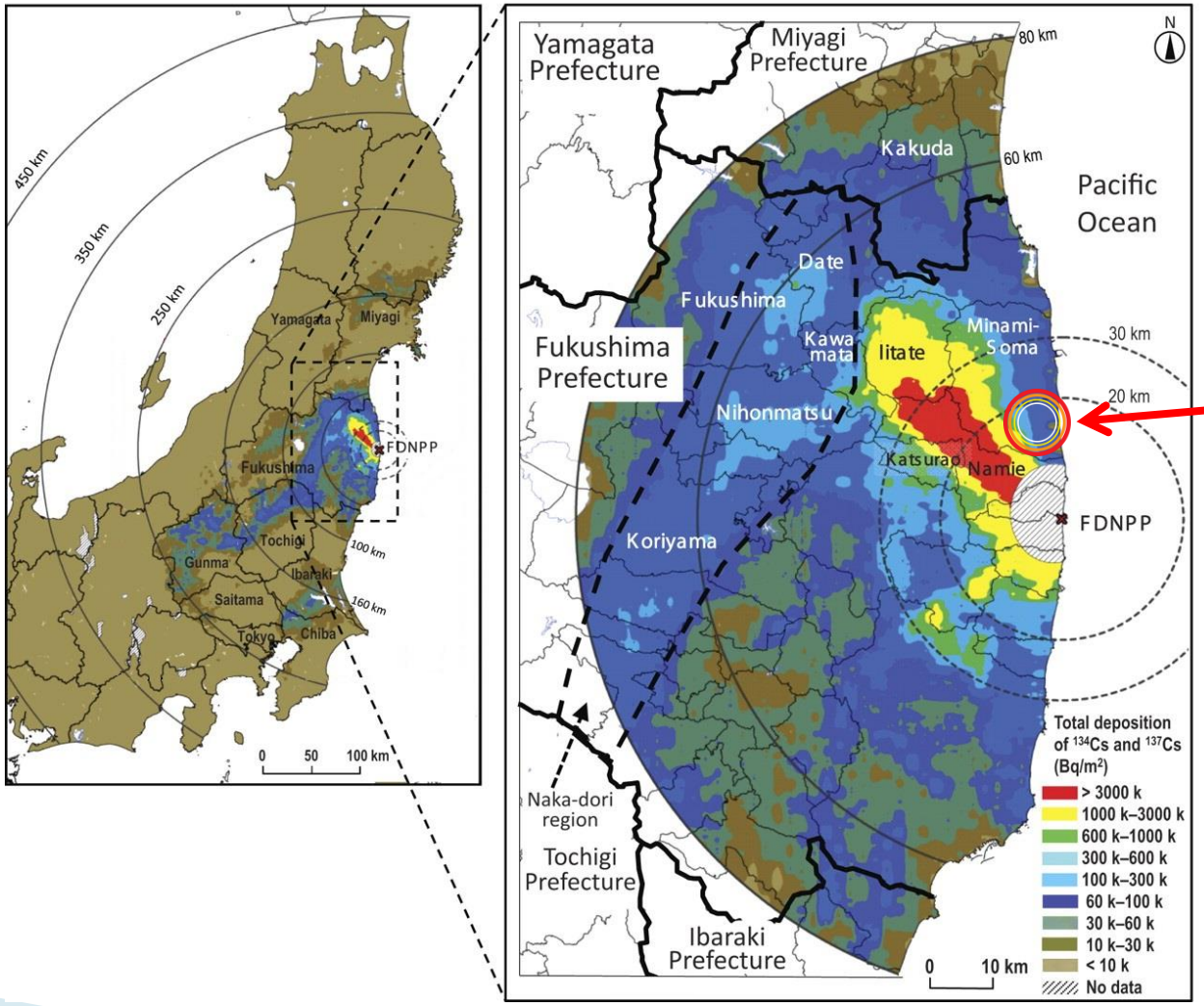


Spot G:
$^{240}Pu/^{239}Pu$ (at) = 0.64±0.37
A (^{239}Pu) = 0.049±0.021 Bq/kg

Spot A:
$^{240}Pu/^{239}Pu$ (at) = 0.381±0.046
A (^{239}Pu) = 0.204±0.015 Bq/kg

S. Schneider,..., G. Steinhauser, Sci. Rep. 3 (2013) 2988

Spot G



Spot G!

Figure modified after
N. Yoshida, Y. Takahashi , Elements 8 (2012) 201–6

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ENVIRONMENTAL MONITORING
MASS SPECTROMETRY

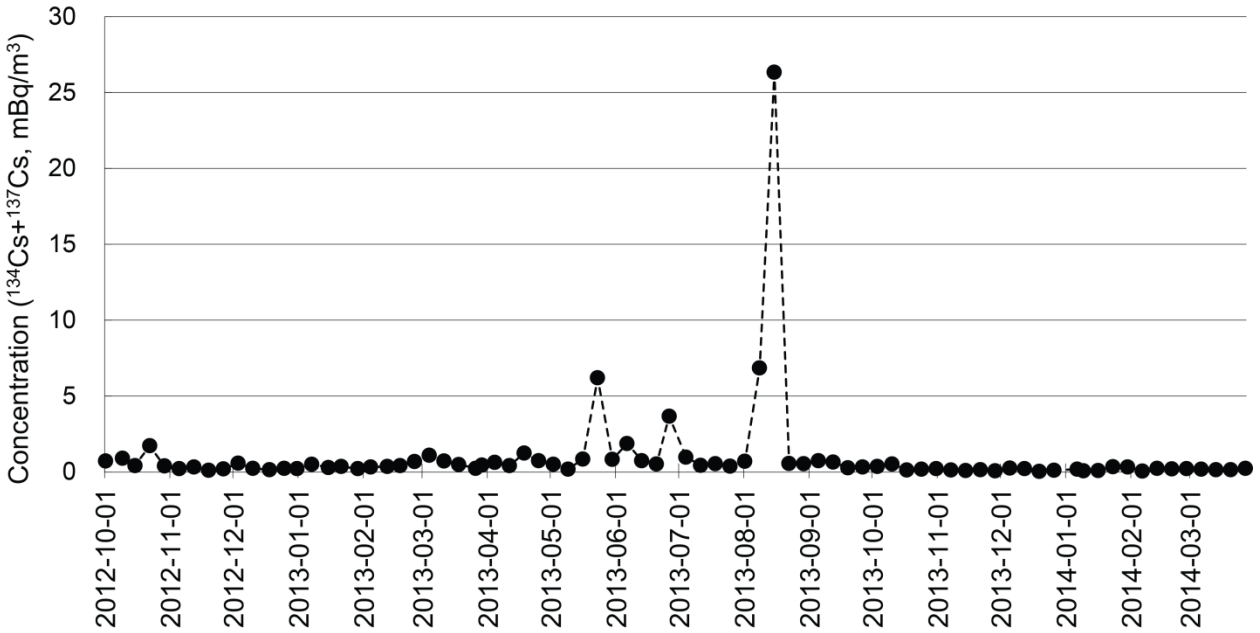
Plutonium release from Fukushima Daiichi fosters the need for more detailed investigations

Stephanie Schneider[1], Clemens Walther[1], Stefan Bister[1], Viktoria Schauer[2], Marcus Christl[3], Hans-Arno Synal[3], Katsumi Shozugawa[4] & Georg Steinhauser[5]

This observation makes it likely that the release of plutonium was a more singular event, whereas the volatile radionuclides were released from the pressure vessels over several days in the course of the early venting operations.

Late radionuclide outbursts?

- Weekly analysis of air filters N, W, and S of the NPP

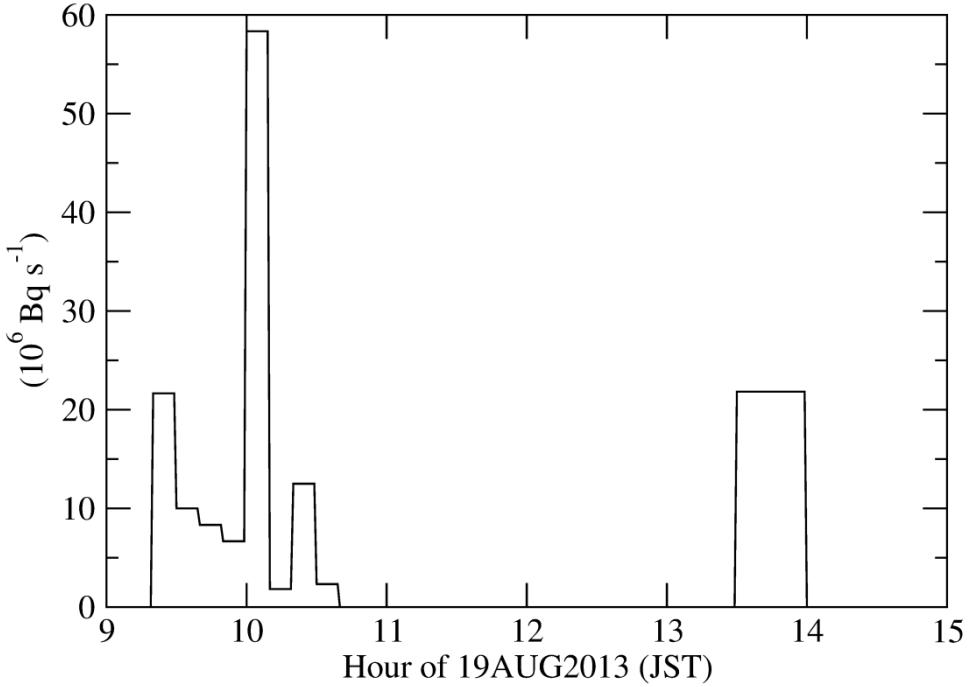


G. Steinhauser et al., Environ. Sci. Technol. 49 (2015) 14028

Hypothesis

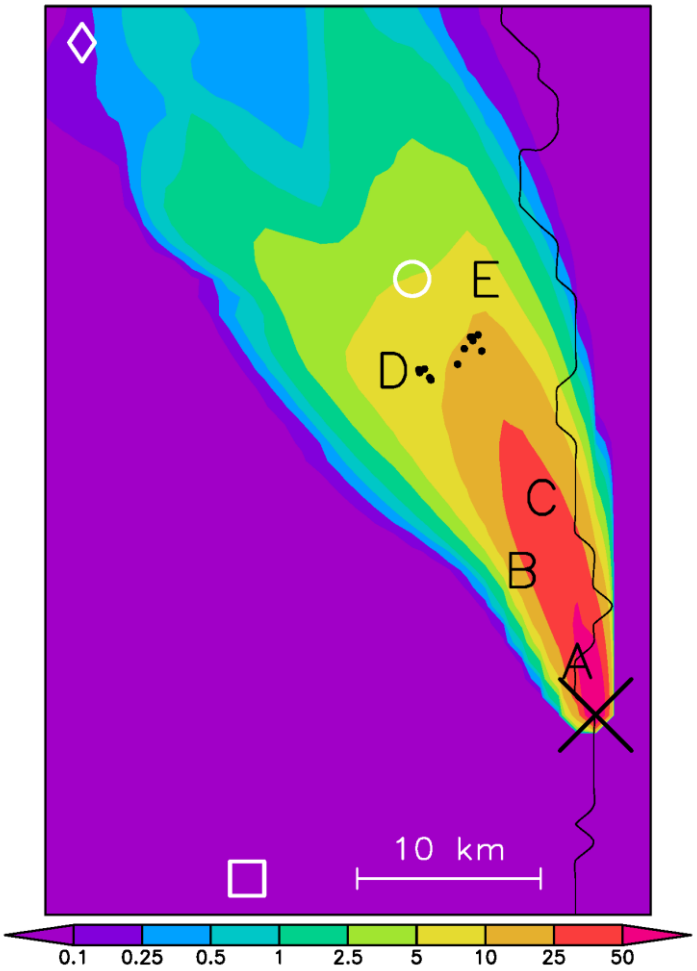
- Late releases caused by debris removal operations (earthmoving operations)?

- Checking TEPCO‘s press releases: Workers complained about having to work in radioactive dust during this particular week.



- Cesium-137 emissions on August 19, 2013 used in the model (10^6 Bq s^{-1}). The emission values were derived from radioactivity estimated by NRA assuming emitted radionuclides were only ^{134}Cs and ^{137}Cs. The total emission of ^{137}Cs was 7.7×10^{11} Bq.

Was it really from the NPP site?



- Searching for indicators of a release from the NPP site:
- Refractory elements!

G. Steinhauser et al., Environ. Sci. Technol. 49 (2015) 14028

Was it really from the NPP site?

- Unusually high ^{90}Sr concentration in only one spot: 78 ± 8 Bq kg^{-1} ^{90}Sr
- $^{90}Sr/^{137}Cs$ activity ratio: 0.04 (very high)

G. Steinhauser et al., Environ. Sci. Technol. 49 (2015) 14028

Thank you for your attention

“We shall return”
(close to the exclusion zone, 7.5 km SW of Fukushima Daiichi NPP)

Courtesy of P. Bossew, BfS

Hydrogen explosions

- Reaction of vapor with Zircaloy cladding:
  $Zr + 2\ H_2O \rightarrow ZrO_2 + 2\ H_2\uparrow$
- Reactor Unit 1: 12 March, 15:31
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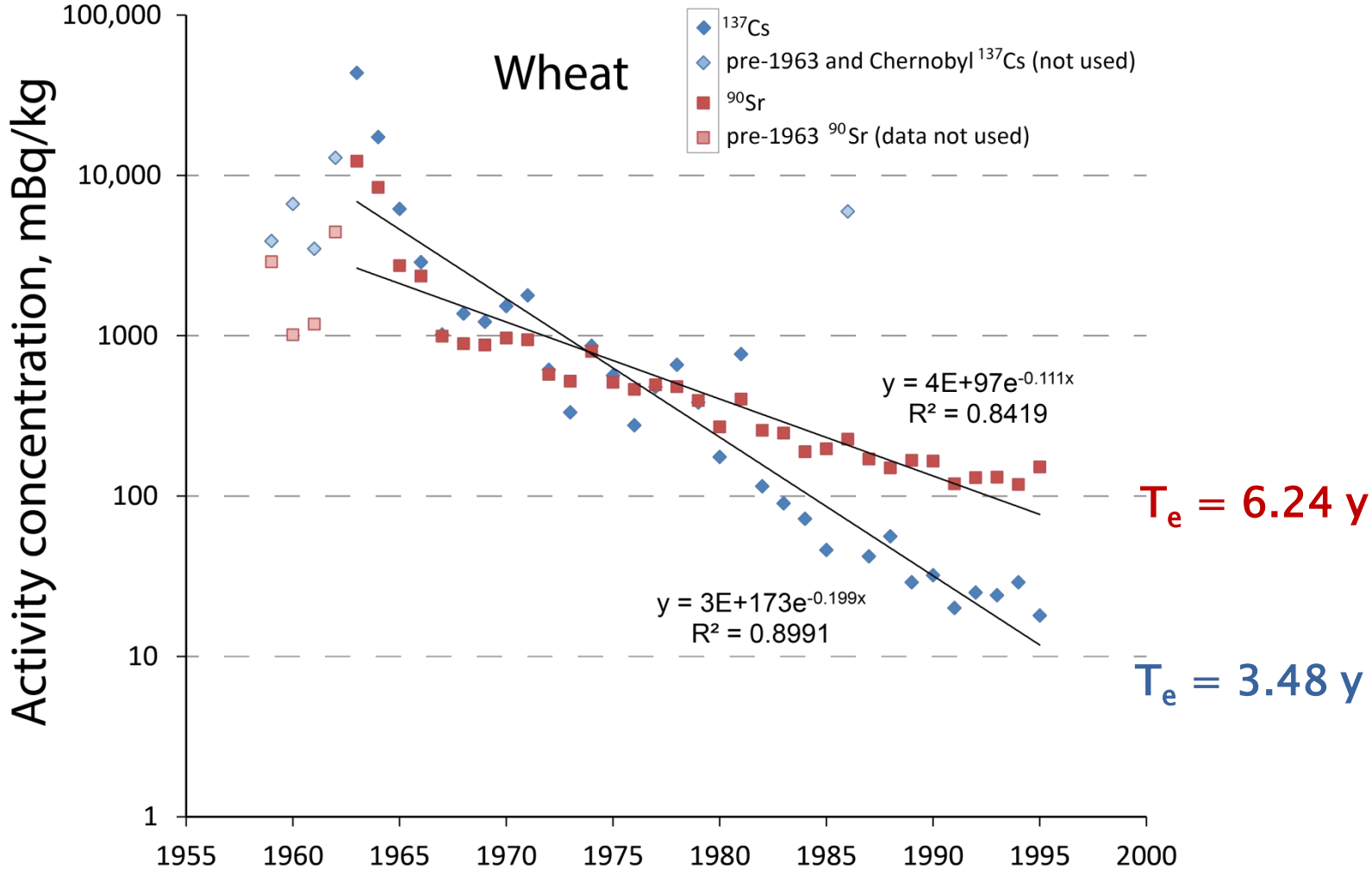
Effective half-lives



Effective half-lives