中国科学: 地球科学 2015 年 第 45 卷 第 12 期: 1875 ~ 1885
www.scichina.com earth.scichina.com
《中国科学》杂志社
SCIENCE CHINA PRESS

论 文

福岛核事故源项评价

林武辉[①②], 陈立奇[①③*], 余雯[①②], 马豪[④], 曾志[④], 曾实[④]

① 国家海洋局第三海洋研究所, 厦门 361005;
② 国家海洋局第三海洋研究所, 海洋放射性技术与环境安全评估实验室, 厦门 361005;
③ 国家海洋局海洋-大气化学与全球变化重点实验室, 厦门 361005;
④ 清华大学工程物理系, 北京 100084
* 联系人, E-mail: chenliqi@tio.org.cn

收稿日期: 2014-12-23; 接受日期: 2015-07-07; 网络版发表日期: 2015-11-20
中国极地科学战略研究基金项目(编号: 20120316)、清华大学自主科研计划项目(编号: 20111080965)和国家自然科学基金项目(批准号: 11205094, 41106167)资助

摘要 2011 年日本福岛发生的核泄漏对全球环境和社会产生巨大影响, 核事故的后果评价与环境修复成为长期而复杂的任务, 而源项作为事故分级的重要依据, 是后果评价和环境修复的基础. 本文从福岛核事故泄漏的核素总量与组成、特征核素谱与其他事件的历史排放水平对比等三方面出发, 对福岛核事故的源项进行分析. 分析结果表明, 福岛核事故泄漏放射性总量大约是切尔诺贝利核事故泄漏总量的 10%, 不到全球落下灰总量的千分之一, 但是福岛核事故是至今为止最为严重的海洋放射性污染核事故. 本文还对核事故后处理措施进行评估, 针对福岛核事故的环境影响提出一些建议.

关键词
福岛核事故
切尔诺贝利核事故
全球落下灰
放射性
大气
海洋

2011年3月11号日本东部海域发生9级大地震, 随后引发的海啸浪高超过14 m, 破坏福岛第一核电站的冷却系统, 并随即引发堆芯融化、氢气爆炸, 大量放射性物质泄漏进入大气、海洋和陆地环境(Povinec等, 2013c).

日本福岛核事故被国际原子能机构(IAEA)定为7级核事故, 其泄漏放射性物质总量大约为切尔诺贝利核事故的10%~15%(Steinhauser等, 2014), 却是至今为止最严重的海洋放射性污染的核事故(Povinec等, 2013c; Buesseler, 2014). 福岛核事故排放的放射性物质影响整个北半球(Thakur等, 2013), 甚至部分南半球地区(Orr等, 2013).

核事故后的源项估计是事故分级的重要依据, 是后果评价的基础. 福岛核事故放射性物质排放存在间断式的事件排放与长时期非点源排放. 福岛核事故之后由于电力与通信的缺失, 核事故初期监测数据的空白, 核事故后期存在一些不可预期的非点源排放, 同时监测站位与频率的限制等, 都给源项估计带来很大挑战.

福岛核事故已过去3年, 国际上已经开展大量的研究, 但较为分散. 本文从源项入手, 大量收集福岛核事故泄漏的放射性物质经大气、海洋途径排放的源项数据; 从历史排放角度出发, 对比福岛核事故、切尔诺贝利核事故、全球落下灰的排放量; 从事故排放的放射性核素组成出发, 寻找日本福岛核事故的指纹特征, 便于今后端元分析, 定量不同源项的核素贡献;

中文引用格式: 林武辉, 陈立奇, 余雯, 马豪, 曾志, 曾实. 2015. 福岛核事故源项评价. 中国科学: 地球科学, 45: 1875–1885
英文引用格式: Lin W H, Chen L Q, Yu W, Ma H, Zeng Z, Zeng S. 2015. Radioactive source-term of the Fukushima Nuclear Accident. Science China: Earth Sciences, doi: 10.1007/s11430-015-5112-8

对同为7级的福岛核事故与切尔诺贝利核事故进行比较; 最后针对福岛核事故的源项进行总结及展望.

1 大气排放

核事故后的大气排放是对环境影响最为直接、迅速, 影响范围最为广泛的途径. 福岛核事故后许多研究者开展大气排放放射性物质总量的评估(Bailly du Bois等, 2014; Cervone和Franzese, 2014; Koo等, 2014; Lin等, 2015b), 但是仍然存在部分核素未被统计. 全面了解福岛核事故来源的人工放射性核素排放的总量与组成, 对于事故分析、辐射评价、环境修复和核素生物地球化学过程研究等具有重要意义(Steinhauser, 2014). 表1给出至今为止最为全面的不同核素通过大气排放的放射性总量估计, 由于日本方面发布信息透明度及可信度存疑, 监测站位与频率的不足, 源项估计时间、空间及方法上的差异等, 不同研究者对于同一核素排放总量的估算存在差异, 本文一并保留并给出相应出处, 以便今后进一步深入的分析与评价.

^{131}I, ^{134}Cs和^{137}Cs由于其分析方法相对简单, 核事故后有大量的环境监测数据, 同时三者也是辐射剂量评价中最为主要的核素, 因此对于三者的源项估计研究最多. 从表1中可以得到^{131}I, ^{134}Cs和^{137}Cs大气排放的源项分别为60~390, 15~20和5~50 PBq. 不同大气输运模型结构差异, 也将影响源项的估算结果.

2 海洋排放

福岛核事故泄漏大量放射性物质直接进入海洋, 是至今为止最为严重的海洋放射性污染的核事故(Lin等, 2015a). 冷却水与堆芯直接接触, 除^{131}I, ^{134}Cs和^{137}Cs等低沸点核素外, 部分高沸点的^{90}Sr和$^{239+240}$Pu核素也可能泄露进入海洋环境(Povinec等, 2012; Bu等, 2013a; Casacuberta等, 2013; Zheng等, 2013), 核素泄漏总量与成分取决于冷却水酸度与液体排放量. 表2总结目前最为全面的不同核素通过海洋途径排放的放射性源项估计.

由于海洋监测复杂性, 海洋排放的研究相对于大气排放的研究较少. 陆地环境中, 通过大气排放的U, Pu, Am和Cm等核素沉降于核电站附近的土壤(Yamamoto等, 2012, 2014a, 2014b; Zheng等, 2012; Schneider等, 2013; Sakaguchi等, 2014)、植物表面(Shozugawa等, 2012; Schneider等, 2013)和河流沉积物(Evrard等, 2014)和气溶胶(Shinonaga等, 2014)等, 来自福岛核事故的核素可以被采样、测量、识别, 表1根据陆地环境测量结果, 反推福岛核事故来源的超铀元素的泄漏量; 海洋环境中也必然存在福岛核事故来源的U, Pu, Am和Cm等核素, 然而由于海洋较强的稀释能力, 福岛核事故排放的放射性物质进入海洋后迅速被稀释, 至今为止海洋环境中还没有来自福岛核事故的超铀核素的直接证据(Bu等, 2013a, 2013b, 2014a, 2014b; Oikawa等, 2014), 因此表2并未给出海洋途径排放的超铀元素的泄漏量.

^{131}I, ^{134}Cs和^{137}Cs作为海洋辐射剂量评价关注的焦点核素, 根据表2得到, ^{131}I, ^{134}Cs和^{137}Cs海洋排放的源项分别为10~61, 2~20和1~30 PBq. ^{137}Cs作为一个长半衰期的重要人工放射性核素, 其研究结果最为丰富, Buesseler等(2012)的结果是西北太平洋调查海域的^{137}Cs储量, 因此2 PBq是非常保守的估计; 东京电力公司(TEPCO)基于实测估算2号反应堆中高放射性冷却水中^{137}Cs的泄漏量为4.7 PBq, 该结果尚未考虑1~4号反应堆的泄露, 地下水排放, 人为低放射性废水排放等过程, 因此该结果也较为保守; Charette等(2013)根据Buesseler等(2012)的实测结果, 结合镭同位素数据估算海洋途径^{137}Cs排放量为11~16 PBq, 该结果通过镭获得物理输运过程的参数, 由于数据稀缺, 存在较大的不确定度; Bailly du Bois等(2012)估算的^{137}Cs通过海洋途径的排放总量为27 PBq, 该结果考虑了更多过程的海洋输入途径. 根据以上分析及表2结果, 作者认为福岛核事故来源的^{137}Cs的海洋途径排放量保守估计为5 PBq.

虽然福岛核事故后, 多个放射性物质输运模型曾经进行比较(Masumoto等, 2012), 不同模型的不确定性来源存在差异, 而且程度也不同, 某一个模型的结果很难全面代表源项情况, 因此源项评价是一个长期过程, 需要利用不同技术、从不同角度开展工作, 减少不确定度, 增加结果的可信度.

3 源项对比

福岛核事故与切尔诺贝利核事故都被IAEA定为最高的7级核事故, 为更好的了解福岛核事故源项排

表 1　大气排放的放射性物质源项估计(10^{15} Bq)

核素	排放总量
^{85}Kr	44.1(Ahlswede等, 2013) 61(Nitta等, 2014) 83.7(Povinec等, 2013c)
^{90}Sr	0.14(Povinec等, 2013c)
^{110m}Ag	1.5×10^{-2}~0.15(Saito等, 2014), 3.0×10^{-2}(Lepage等, 2014)
^{129m}Te	3.33(Povinec等, 2013c)
^{129}I	8.06×10^{-6}(Hou等, 2013)
^{131}I	65.3(Ten Hoeve和Jacobson, 2012), 105.9(Saunier等, 2013) 120(Nagai等, 2014), 100~400(Achim等, 2012) 151(Katata等, 2012), 153~160(Chino等, 2011) 159(Povinec等, 2013c), 160(Povinec等, 2013c) 190(Mathieu等, 2012), 100~200(Akahane等, 2012) 190~380(Winiarek等, 2012), 320(TEPCO, 2011) 200(IRSN, 2011; Kobayashi等, 2013) 360~390(ZAMG 2011)
^{132}I	35.8(Saunier等, 2013)
^{132}Te	88(Tagami等, 2013), 88.4(Povinec等, 2013c)
^{133}I	42.2(Povinec等, 2013c)
^{133}Xe	6000(Achim等, 2012), 12000(Bowyer等, 2011; Povinec等, 2013c) 22000(TEPCO, 2011), 12134(Saunier等, 2013) 14000(Stohl等, 2012a), 15300(Stohl等, 2012) 17000(NILU, 2011), 20000(IRSN, 2011)
^{134}Cs	15(Povinec等, 2013c), 17.5(Povinec等, 2013a) 18(Hamada和Ogino, 2012)
^{135}Cs	6.74×10^{-5}(Zheng等, 2014)
^{137}Cs	5.5~9.7(Miyazawa等, 2012), 8.8(Nagai等, 2014) 11.5(Bailly du Bois等, 2012), 10(Morino等, 2011; Achim等, 2012) 12(Yasunari等, 2011; Winiarek等, 2012) 13(Kawamura等, 2011; Kobayashi等, 2013) 15(Honda等, 2012; Povinec等, 2013c) 15.3(Povinec等, 2013b), 15.5(Saunier等, 2013) 13~15(Chino等, 2011), 17(Ten Hoeve和Jacobson, 2012) 20(Mathieu等, 2012; Korsakissok等, 2013; Bailly du Bois等, 2014) 10~20(Akahane等, 2012) 30(IRSN, 2011), 36(NILU, 2011), 36.6(Stohl等, 2012b) 50(ZAMG (Central Institue for Meteorology and Geodynamics, 2011)
U	3.9×10^{-9}(Sakaguchi等, 2014)
$^{239+240}$Pu	1×10^{-6}~2.4×10^{-6}(Zheng等, 2012), 2.3×10^{-6}(Sakaguchi等, 2014) 3.5×10^{-6}(Yamamoto等, 2014b)
^{241}Am	5.0×10^{-7}~1.3×10^{-6}(Yamamoto等, 2014b)
^{242}Cm	2.7×10^{-5}~6.5×10^{-5}(Yamamoto等, 2014b)
$^{243+244}$Cm	1.1×10^{-6}~2.6×10^{-6}(Yamamoto等, 2014b)

表 2　海洋排放的放射性物质源项估计(10^{15} Bq)

核素	排放总量
^{3}H	0.1(Povinec等, 2013c)
^{90}Sr	0.09~0.9(Casacuberta等, 2013) 0.08(Periáñez等, 2013), 1~6(Povinec等, 2012)
^{99m}Tc	0.02(Nair等, 2014)
^{129}I	2.35×10^{-6}(Hou等, 2013) 7×10^{-6}(Guilderson等, 2013; Povinec等, 2013a)
^{131}I	11(Kawamura等, 2011), 11.1(Tsumune等, 2013) 61.6(Hou等, 2013)
^{134}Cs	2.24(Nair等, 2014), 3.5(Tsumune等, 2013) 11~16(Charette等, 2013)
^{135}Cs	7.01×10^{-5}(Zheng等, 2014)
^{136}Cs	0.29(Nair等, 2014)
^{137}Cs	0.9~3.5(Dietze和Kriest, 2012), 2(Buesseler等, 2012) 2.2(Povinec等, 2013c), 3.5(Tsumune等, 2012, 2013) 3.6(Tsumune等, 2013), 4(Kawamura等, 2011) 5.1~5.5(Estournel等, 2012), 5.5~5.9(Miyazawa等, 2012) 16.2(Rypina等, 2013), 27(Bailly du Bois等, 2012)
^{140}Ba	0.53(Nair等, 2014)
^{140}La	0.27(Nair等, 2014)

放情况, 表3给出全球落下灰、切尔诺贝利核事故、福岛核事故排放的放射性物质对比情况, 便于从历史排放水平的角度上, 更直观的了解福岛核事故的源项排放情况.

根据表3可知, 如果不考虑放射性惰性气体的排放(^{85}Kr和^{133}Xe), 福岛核事故来源的^{131}I和^{137}Cs等主要的放射性核素排放量总量大约为切尔诺贝利核事故的10%, 不足全球落下灰的千分之一; 如果考虑放射性惰性气体的排放(^{85}Kr和^{133}Xe), 福岛核事故泄漏放射性物质总量高于切尔诺贝利核事故, 因为福岛核事故共有4个反应堆发生不同程度的堆芯熔化, 而切尔诺贝利核事故只有一个反应堆发生堆芯熔化; 对于$^{239+240}$Pu, 福岛核事故泄漏量远小于切尔诺贝利核事故和全球落下灰的排放量.

4　福岛核事故排放的核素谱的特征指纹

不同核事故泄漏的特征核素谱存在差异, 取决于核燃料组成、反应堆燃耗、泄漏过程和冷却时间等因素. 了解事故后核素活度比值与原子数比值, 可以判断污染源、反演历史排放情况、估算其他核素的排放总量、了解堆芯融化情况、评估核燃料燃耗等. 对于同一污染源, 核素运移过程中存在生物地球化学行为差异、同位素分馏、不同源项的混合、放射衰变等过程, 将导致核素比值的改变; 对于不同污染源, 核燃料组成、反应堆燃耗情况、核燃料泄漏过程及方式、核燃料冷却时间等将影响核素比值. 表4对比日本福岛核事故、切尔诺贝利核事故、全球落下灰核素比值, 该表有利于今后不同端元的确定, 计算不同来源放射性物质含量的贡献.

核素活度比值、原子数比值还可以用来估计其他核素的排放总量, 特别是对于一些分析方法较为复杂的核素(^{90}Sr和^{129}I), 事故之后由于测量资源的限制(时间、仪器、人力), 无法同时大量开展监测, 可以

表 3　福岛核事故、切尔诺贝利核事故与全球落下灰放射性物质排放量(10^{15} Bq)

	福岛核事故		切尔诺贝利核事故		全球落下灰	
	大气	海洋	大气	海洋	大气	海洋
^{131}I	160[a)]	11[h)]	1760[j)]			
^{137}Cs	15[a)]	4[h)]	85[j)]	16[m)]	950[j)]	600[m)]
^{90}Sr	0.14[b)]	1[i)]	10[j)]		600[j)]	380[m)]
^{133}Xe	1.2×10^{4}~1.5×10^{4}[c),d)]		2.0×10^{3}~6.0×10^{3}[k)]			
^{85}Kr	44.1[e)]		33[e)]		5000	
$^{239+240}$Pu	1.0×10^{-6}~2.4×10^{-6}[f)]		0.1[l)]		10[j)]	
排放总量[n)]	520[g)]		5200[l)]		712920[j)]	

a) Povinec等(2013b); b) Povinec等(2013c); c) Bowyer等(2011); d) Stohl等(2012b); e) Ahlswede等(2013); f) Zheng等(2012); g) Steinhauser等(2014); h) Kawamura等(2011); i) Povinec等(2012); j) UNSEAR(2008); k) Ginzburg和Reis(1991); l) Livingston和Povinec(2002); m) IAEA(2005); n) 排放总量中不包含放射性惰性气体(^{85}Kr和^{133}Xe)

表 4 福岛核事故、切尔诺贝利核事故和全球落下灰核素比值

	福岛核事故		切尔诺贝利核事故		全球落下灰	
	活度比值	原子数比值	活度比值	原子数比值	活度比值	原子数比值
$^{134}Cs/^{137}Cs$	1[a)~c)]		0.5[a)]			
$^{135}Cs/^{137}Cs$[d)~f)]		0.37		0.46		2.7
$^{131}I/^{137}Cs$	15[g)]		20			
$^{129}I/^{131}I$		22.3[h)]		19[i)]		
$^{129}I/^{137}Cs$[j)]	4.43×10^{-7}		2.75×10^{-7}			
$^{90}Sr/^{137}Cs$	0.01~0.02[b),k)]		0.08[l)]		0.63[b)]	
$^{240}Pu/^{239}Pu$[m)]		0.3		0.4		0.18
$^{241}Pu/^{239}Pu$		0.103~0.135[m)]				1.9×10^{-3}[n)]
$^{241}Pu/^{239+240}Pu$	107.8[m)]		83[m)]		13~14[o)]	
$^{238}Pu/^{239+240}Pu$	2.2[p)]		0.5[o)]		0.033[p)]	

a) Masson等(2011); b) Buesseler等(2012); c) Merz等(2013); d) Ohno和Muramatsu(2014); e) Shibahara等(2014); f) Zheng等(2014); g) Hirose(2012); h) Miyake等(2012); i) Kutschera等(1988); j) Tumey等(2013); k) Periáñez等(2013); l) Aarkrog(2003); m) Zheng等(20120; n) Kelley等(1999); o) Hirose等(2001); p) Lujanienė等(2012)

通过该核素与其他容易测量的核素(^{137}Cs和^{131}I)比值间接估算事故中的泄漏量; 同时核素活度和原子数比值还可以用来判断放射性物质的来源, 定量不同来源的源项贡献.

5 福岛核事故与切尔诺贝利核事故比较

福岛核事故与切尔诺贝利核事故的差异从三方面展开讨论: 放射性物质总量、放射性物质成分和核素迁移途径.

对于泄漏放射性物质总量, 虽然日本福岛核事故有4个反应堆发生氢气爆炸, 并伴随着不同程度的放射性物质泄漏, 切尔诺贝利核事故只有1个反应堆发生事故, 在不考虑惰性气体前提下, 福岛核事故泄漏放射性物质总量为520 PBq, 大约为切尔诺贝利核事故总量的10%~15%(Steinhauser等, 2014), 如果考虑放射性惰性气体(^{85}Kr和^{133}Xe), 福岛核事故泄漏放射性物质总量大于切尔诺贝利核事故.

对于泄漏放射性物质成分, Nishihara等(2012)对福岛核电站反应堆、乏燃料中的放射性核素清单进行计算, 详细给出反应堆内16种裂变核素、10种锕系核素、2种活化元素活度总量(Nishihara等, 2012); Yamamoto在福岛核电站周边土壤测得Sr, Nb, Mo, Tc, Ru, Ag, Te, I, Ba, La, Pu, Am和Cm的同位素, 并根据$^{134}Cs/^{137}Cs$、$^{238}Pu/^{239+240}Pu$比值推测反应堆的燃耗情况, 估算其他核素与^{137}Cs的比值(Yamamoto, 2012); 福岛核电站周边的树木和碎石中也检测到^{3}H, ^{14}C, ^{60}Co, ^{79}Se, ^{90}Sr, ^{99}Tc和^{137}Cs等核素(Tanaka等, 2014); Kirchner等(2012)认为事故时刻堆芯温度小于2700 K(Kirchner等, 2012), 福岛核事故泄漏放射性核素主要是一些沸点较低的易挥发性核素, 比如: 惰性气体(^{85}Kr和^{133}Xe)(Nitta等, 2014), ^{95}Zr-^{95}Nb(Tagami等, 2011; Kanai, 2012), ^{99}Mo-^{99m}Tc(Kanai, 2012), ^{110m}Ag(Kanai, 2012; Saegusa等, 2013; Lepage等, 2014; Saito等, 2014), ^{125}Sb(Saegusa等, 2013), ^{129}I(Miyake等, 2012), ^{131}I(Chino等, 2011), ^{133}I(Leon等, 2011), $^{134,135,136,137}Cs$(Tagami等, 2011; Kanai, 2012; de Vismes Ott等, 2013; Zheng等, 2014), ^{129m}Te-^{129}Te(Kanai, 2012; de Vismes Ott等, 2013), ^{132}Te-^{132}I(Tagami等, 2011; Kanai, 2012; de Vismes Ott等, 2013; Tagami等, 2013), ^{140}Ba-^{140}La(Tagami等, 2011; Kanai, 2012; de Vismes Ott等, 2013)等, 而沸点较高的核素, 比如^{59}Fe, ^{90}Sr, ^{95}Zr, ^{236}U及^{239}Np, $^{239,240}Pu$, Am和Cm等超铀核素泄漏量较少(Tagami等, 2011; Shozugawa等, 2012; Zheng等, 2012; Schneider等, 2013; Steinhauser等, 2013; Zheng等, 2013; Sakaguchi等, 2014; Yamamoto等, 2014a), 大部分留在反应堆内部(Schwantes等, 2012). 但是部分研究表明事故期间日本气溶胶中仍然可以检测到福岛核事故产生的$^{239+240}Pu$, ^{236}U(Shinonaga等, 2014), 欧洲检测气溶胶中$^{239+240}Pu$在福岛核事故期间轻微升

高, 但是无法肯定是否来自福岛核事故(Lujanienė等, 2012). Tanabe(2012), Schwantes等(2012)和Le Petit等(2012)根据环境中核素的测量数据对堆芯情况与放射性核素泄漏进行分析(Blandford和Ahn, 2012; Le Petit等, 2012; Schwantes等, 2012; Tanabe, 2012).

因此, 福岛核事故主要释放易挥发的放射性核素(放射性惰性气体, I, Cs和Te), 而高沸点的核素(Sr, 锕系核素, Zr和Ce等)只有在核电站周边地区发现(Doi等, 2013), 日本以外的北美、欧亚大陆等并未发现高沸点的核素, 而切尔诺贝利核事故除了泄漏挥发性核素外, 还包含大量的Sr, Ru, Zr, Ce, La, Pm和Pu等高沸点核素(Kashparov等, 2003).

福岛核事故排放的放射性物质80%进入太平洋, 19%沉降于日本, 不到1%沉降于北美、欧亚大陆等地区(Morino等, 2011; Stohl等, 2012b; Ten Hoeve和Jacobson, 2012; Yoshida和Kanda, 2012; Christoudias和Lelieveld, 2013), 主要污染北太平洋; 切尔诺贝利核事故只有不到10%量沉降于海洋且主要污染波罗的海、黑海等北欧海域, 大部分放射性物质沉降于欧洲大陆(Evangeliou等, 2014). 由于日本独特的物理环境, 大部分放射性物质进入海洋, 福岛核事故对陆地污染范围较切尔诺贝利核事故的污染范围小(Steinhauser等, 2014), 但是海洋环境的影响范围较大.

6 结论与展望

不考虑惰性气体排放, 福岛核事故排放放射性物质总量大约为520 PBq, 是切尔诺贝利核事故的10%左右, 不足全球落下灰的千分之一; 福岛核事故泄漏的放射性物质分布于日本、太平洋和世界其他地区的比重大约为19%, 80%和1%. 因此切尔诺贝利核事故仍然是至今为止最为严重的核事故, 而福岛核事故是至今为止最为严重的海洋放射性污染核事故.

福岛核事故核素排放组成以低沸点的Kr, Xe, I, Cs和Te为主, 高沸点Sr和Pu等核素大部分停留于反应堆内部, 福岛核事故源项具有独特的放射性核素谱, 与全球落下灰、切尔诺贝利核事故的核素谱存在差异, 核素活度或者原子数比值今后可作为一个典型的历史事件而记录于环境介质中. 日本附近的湖泊和海洋沉积物、陆地土壤中, 来自福岛核事故的^{137}Cs很可能可以作为今后独特的历史事件而被识别, 并应用于地质年代学研究中, 但是世界其他地区的沉积物中该事件将很难被识别.

目前对核事故中燃料棒、控制棒、压力壳和冷却剂等物理、化学等过程的了解还不充分, 特别是淡水、海水与核燃料直接接触, 放射性物质液态排放的总量与组成, 核电站周边的非点源长期排放等仍然存在很大不确定性(Burns等, 2012). 事故后人为引入大量的冷却水与熔化后的核燃料仍然停留于核电站内部, 并可能通过地下水进入环境中. 河流输入也可能作为一个长期的排放途径, 把沉降于日本陆地的放射性物质搬运至近海海域. 核电站附近储存高放射性废水的废液罐也是潜在的污染源. 目前核电站附近海域放射性活度仍然居高不下, 说明仍然存在持续的泄漏源项(Kanda, 2013). 寻找泄漏途径、定量泄漏通量、有效控制泄漏总量是日本今后一项长期而又艰巨的任务.

核事故后采取的冷却措施导致核电站周边储存着大量的高放射性的冷却水, 日本联合美国、俄罗斯等国家正积极开展高放射性废液后处理的研究(Sylvester等, 2013), 许多核素可以通过化学方法去除, 但是^{3}H无法通过化学方法去除, 最终也很可能直接排入海洋, 其他利益相关方或者国际组织应该及时建立环太平洋^{3}H本底, 以此关注日本方面采取的冷却水处理措施.

核事故的后果评价与环境修复是一项长期而复杂的任务, 而源项的准确估计是最为基础的工作. 尽管国际上已经开展大量的研究工作, 但是不同排放时间及空间的界定、不同来源的气象与水文数据的选取、不同模型的应用、不同来源的监测数据的校准等因素, 都将导致福岛核事故的源项估计仍然存在很大不确定性. 今后仍然需要利用更多手段, 从不同角度出发定量源项, 减少源项评估过程中的不确定度, 增加源项评估结果的可信度.

参考文献

Aarkrog A. 2003. Input of anthropogenic radionuclides into the World Ocean. Deep-Sea Res Part II-Top Stud Oceanogr, 50: 2597–2606

Achim P, Monfort M, Le Petit G, et al. 2012. Analysis of radionuclide releases from the Fukushima Dai-ichi nuclear power plant accident part II. Pure Appl Geophys, 171: 645–667

Ahlswede J, Hebel S, Ross J O, et al. 2013. Update and improvement of the global krypton-85 emission inventory. J Environ Radioact, 115: 34–42

Akahane K, Yonai S, Fukuda S, et al. 2012. The Fukushima Nuclear Power Plant accident and exposures in the environment. Environmentalist, 32: 136–143

Bailly du Bois P, Laguionie P, Boust D, et al. 2012. Estimation of marine source-term following Fukushima Dai-ichi accident. J Environ Radioact, 114: 2–9

Bailly du Bois P B, Garreau P, Laguionie P, et al. 2014. Comparison between modelling and measurement of marine dispersion, environmental half-time and ^{137}Cs inventories after the Fukushima Daiichi accident. Ocean Dynam, 64: 361–383

Blandford E D, Ahn J. 2012. Examining the nuclear accident at Fukushima Daiichi. Elements, 8: 189–194

Bowyer T W, Biegalski S R, Cooper M, et al. 2011. Elevated radioxenon detected remotely following the Fukushima nuclear accident. J Environ Radioact, 102: 681–687

Bu W, Fukuda M, Zheng J, et al. 2014a. Release of Pu isotopes from the Fukushima Daiichi Nuclear Power Plant accident to the marine environment was negligible. Environ Sci Technol, 48: 9070–9078

Bu W, Zheng J, Aono T, et al. 2013a. Vertical distributions of plutonium isotopes in marine sediment cores off the Fukushima coast after the Fukushima Dai-ichi Nuclear Power Plant accident. Biogeosciences, 10: 2497–2511

Bu W, Zheng J, Guo Q, et al. 2013b. A method of measurement of ^{239}Pu, ^{240}Pu, ^{241}Pu in high U content marine sediments by sector field ICP-MS and its application to fukushima sediment samples. Environ Sci Technol, 48: 534–541

Bu W, Zheng J, Guo Q, et al. 2014b. Ultra-trace plutonium determination in small volume seawater by sector field inductively coupled plasma mass spectrometry with application to Fukushima seawater samples. J Chromatogr A, 1337: 171–178

Bu W, Zheng J, Guo Q, et al. 2015. Temporal distribution of plutonium isotopes in marine sediments off Fukushima after the Fukushima Dai-ichi Nuclear Power Plant accident. J Radioanal Nucl Chem, 303: 1151–1154

Buesseler K. 2014. Fukushima and ocean radioactivity. Oceanography, 27: 92–105

Buesseler K O, Jayne S R, Fisher N S, et al. 2012. Fukushima-derived radionuclides in the ocean and biota off Japan. Proc Natl Acad Sci USA, 109: 5984–5988

Burns P C, Ewing R C, Navrotsky A. 2012. Nuclear fuel in a reactor accident. Science, 335: 1184–1188

Casacuberta N, Masqué P, Garcia-Orellana J, et al. 2013. ^{90}Sr and ^{89}Sr in seawater off Japan as a consequence of the Fukushima Dai-ichi nuclear accident. Biogeosciences, 10: 3649–3659

Cervone G, Franzese P. 2014. Source term estimation for the 2011 Fukushima nuclear accident. In: Cervone G, Lin J, Waters N, eds. Data Mining for Geoinformatics Methods and Applications. New York: Springer. 49–64

Charette M A, Breier C F, Henderson P B, et al. 2013. Radium-based estimates of cesium isotope transport and total direct ocean discharges from the Fukushima Nuclear Power Plant accident. Biogeosciences, 10: 2159–2167

Chino M, Nakayama H, Nagai H, et al. 2011. Preliminary estimation of release amounts of ^{131}I and ^{137}Cs accidentally discharged from the Fukushima Daiichi nuclear power plant into the atmosphere. J Nucl Sci Technol, 48: 1129–1134

Christoudias T, Lelieveld J. 2013. Modelling the global atmospheric transport and deposition of radionuclides from the Fukushima Dai-ichi nuclear accident. Atmos Chem Phys, 13: 1425–1438

de Vismes Ott A, Gurriaran R, Cagnat X, et al. 2013. Fission product activity ratios measured at trace level over France during the Fukushima accident. J Environ Radioact, 125: 6–16

Dietze H, Kriest I. 2012. ^{137}Cs off Fukushima Dai-ichi, Japan-model based estimates of dilution and fate. Ocean Sci, 8: 319–332

Doi T, Masumoto K, Toyoda A, et al. 2013. Anthropogenic radionuclides in the atmosphere observed at Tsukuba: Characteristics of the radionuclides derived from Fukushima. J Environ Radioact, 122: 55–62

Estournel C, Bosc E, Bocquet M, et al. 2012. Assessment of the amount of cesium-137 released into the Pacific Ocean after the Fukushima accident and analysis of its dispersion in Japanese coastal waters. J Geophys Res, 117: C11014

Evangeliou N, Balkanski Y, Cozic A, et al. 2014. How “lucky” we are that the Fukushima disaster occurred in early spring: Predictions on the contamination levels from various fission products released from the accident and updates on the risk assessment for solid and thyroid cancers. Sci Total Environ, 500: 155–172

Evrard O, Pointurier F, Onda Y, et al. 2014. Novel insights into Fukushima nuclear accident from isotopic evidence of plutonium spread along coastal rivers. Environ Sci Technol, 48: 9334–9340

Ginzburg H, Reis E. 1991. Consequences of the nuclear power plant accident at Chernobyl. Public Health Rep, 106: 32–40

Guilderson T, Tumey S, Brown T, et al. 2013. The 129-Iodine content of subtropical Pacific waters: Impact of Fukushima and other anthropogenic ^{129}I sources. Biogeosciences, 10: 19935–19968

Hamada N, Ogino H. 2012. Food safety regulations: What we learned from the Fukushima nuclear accident. J Environ Radioact, 111: 83–99

Hirose K. 2012. 2011 Fukushima Dai-ichi nuclear power plant accident: Summary of regional radioactive deposition monitoring results. J Environ Radioact, 111: 13–17

Hirose K, Igarashi Y, Aoyama M, et al., 2001. Long-term trends of plutonium fallout observed in Japan. In: A Kudo, ed. Radioactivity in the Environment. New York: Elsevier. 251–266

Honda M C, Aono T, Aoyama M, et al. 2012. Dispersion of artificial caesium-134 and-137 in the western North Pacific one month after the Fukushima accident. Geochem J, 46: 1–9

Hou X, Povinec P P, Zhang L, et al. 2013. Iodine-129 in seawater offshore Fukushima: Distribution, inorganic speciation, sources, and budget. Environ Sci Technol, 47: 3091–3098

IAEA. 2005. Worldwide Marine Radioactivity Studies (WOMARS): Radionuclide Levels in Oceans and Seas. Vienna: IAEA. 187

IRSN (Institute for Radiological Protection and Nuclear Safety). 2011. Simulation of atmospheric dispersion of the radioactive plume formed by releases from the Fukushima Daiichi nuclear power plant since 12 March. http://www.weatheronline.co.uk/daten/weathernews/fukushima/docs/irsn-simulation-dispersion-en.pdf

Kanai Y. 2012. Monitoring of aerosols in Tsukuba after Fukushima Nuclear Power Plant incident in 2011. J Environ Radioact, 111: 33–37

Kanda J. 2013. Continuing ^{137}Cs release to the sea from the Fukushima Dai-ichi Nuclear Power Plant through 2012. Biogeosciences, 10: 6107–6113

Kashparov V, Lundin S, Zvarych S, et al. 2003. Territory contamination with the radionuclides representing the fuel component of Chernobyl fallout. Sci Total Environ, 317: 105–119

Katata G, Ota M, Terada H, et al. 2012. Atmospheric discharge and dispersion of radionuclides during the Fukushima Dai-ichi Nuclear Power Plant accident. Part I: Source term estimation and local-scale atmospheric dispersion in early phase of the accident. J Environ Radioact, 109: 103–113

Kawamura H, Kobayashi T, Furuno A, et al. 2011. Preliminary numerical experiments on oceanic dispersion of ^{131}I and ^{137}Cs discharged into the ocean because of the Fukushima Daiichi nuclear power plant disaster. J Nucl Sci Technol, 48: 1349–1356

Kelley J, Bond L, Beasley T. 1999. Global distribution of Pu isotopes and ^{237}Np. Sci Total Environ, 237: 483–500

Kirchner G, Bossew P, De Cort M. 2012. Radioactivity from Fukushima Dai-ichi in air over Europe; part 2: What can it tell us about the accident ? J Environ Radioact, 114: 35–40

Kobayashi T, Nagai H, Chino M, et al. 2013. Source term estimation of atmospheric release due to the Fukushima Dai-ichi Nuclear Power Plant accident by atmospheric and oceanic dispersion simulations: Fukushima NPP Accident Related. J Nucl Sci Technol, 50: 255–264

Koo Y H, Yang Y S, Song K W. 2014. Radioactivity release from the Fukushima accident and its consequences: A review. Prog Nucl Energy, 74: 61–70

Korsakissok I, Mathieu A, Didier D. 2013. Atmospheric dispersion and ground deposition induced by the Fukushima Nuclear Power Plant accident: A local-scale simulation and sensitivity study. Atmos Environ, 70: 267–279

Kutschera W, Fink D, Paul M, et al. 1988. Measurement of the ^{129}I/^{131}I ratio in Chernobyl fallout. Physica Scripta, 37: 310–313

Le Petit G, Douysset G, Ducros G, et al. 2012. Analysis of radionuclide releases from the Fukushima Dai-Ichi nuclear power plant accident Part I. Pure Appl Geophys, 171: 629–644

Leon J D, Jaffe D, Kaspar J, et al. 2011. Arrival time and magnitude of airborne fission products from the Fukushima, Japan, reactor incident as measured in Seattle, WA, USA. J Environ Radioact, 102: 1032–1038

Lepage H, Evrard O, Onda Y, et al. 2014. Environmental mobility of ^{110m}Ag: Lessons learnt from Fukushima accident (Japan) and potential use for tracking the dispersion of contamination within coastal catchments. J Environ Radioact, 130: 44–55

Lin W, Chen L, He J, et al. 2015a. Review on monitoring marine radioactivity since the Fukushima Nuclear Accident (in Chinese). China Environ Sci, 35: 269–276

Lin W, Chen L, Yu W, et al. 2015b. Radioactivity impacts of the Fukushima Nuclear Accident on the atmosphere. Atmos Environ, 102: 311–322

Livingston H D, Povinec P P. 2002. A millennium perspective on the contribution of global fallout radionuclides to ocean science. Health Physics, 82: 656–668

Lujanienė G, Valiulis D, Byčenkienė S, et al. 2012. Plutonium isotopes and ^{241}Am in the atmosphere of Lithuania: A comparison of different source terms. Atmos Environ, 61: 419–427

Masson O, Baeza A, Bieringer J, et al. 2011. Tracking of airborne radionuclides from the damaged Fukushima Dai-ichi nuclear reactors by European networks. Environ Sci Technol, 45: 7670–7677

Masumoto Y, Miyazawa Y, Tsumune D, et al. 2012. Oceanic dispersion simulations of ^{137}Cs released from the Fukushima Daiichi nuclear power plant. Elements, 8: 207–212

Mathieu A, Korsakissok I, Quélo D, et al. 2012. Atmospheric dispersion and deposition of radionuclides from the Fukushima Daiichi nuclear power plant accident. Elements, 8: 195–200

Merz S, Steinhauser G, Hamada N. 2013. Anthropogenic radionuclides in Japanese food: Environmental and legal implications. Environ Sci Technol, 47: 1248–1256

Miyake Y, Matsuzaki H, Fujiwara T, et al. 2012. Isotopic ratio of radioactive iodine (^{129}I/^{131}I) released from Fukushima Daiichi NPP accident. Geochem J, 46: 327

Miyazawa Y, Masumoto Y, Varlamov S, et al. 2012. Inverse estimation of source parameters of oceanic radioactivity dispersion models associated with the Fukushima accident. Biogeosciences, 9: 13783–13816

Morino Y, Ohara T, Nishizawa M. 2011. Atmospheric behavior, deposition, and budget of radioactive materials from the Fukushima Daiichi nuclear power plant in March 2011. Geophys Res Lett, 38: L00G11

Nagai H, Katata G, Terada H, et al. 2014. Source Term Estimation of ^{131}I and ^{137}Cs Discharged from the Fukushima Daiichi Nuclear Power Plant into the Atmosphere. In: Takahashi S, ed. Radiation Monitoring and Dose Estimation of the Fukushima Nuclear Accident. S. Takahashi: Springer. 155–173

Nair R, Sunny F, Chopra M, et al. 2014. Estimation of radioactive leakages into the Pacific Ocean due to Fukushima nuclear accident. Environ Earth Sci, 71: 1007–1019

NILU. 2011. Flexpart forecast of the atmospheric dispersal of radionuclides from Fukushima. http://flexpart.eu/search?q=fukushima

Nishihara K, Iwamoto H, Suyama K. 2012. Estimation of fuel compositions in Fukushima-Daiichi Nuclear Power Plant. Tokai, Japan Atomic Envergy Agency

Nitta W, Sanada T, Isogai K, et al. 2014. Atmospheric ^{85}Kr and ^{133}Xe activity concentrations at locations across Japan following the Fukushima Dai-ichi Nuclear Power Plant accident. J Nucl Sci Technol, 51: 712–715

Ohno T, Muramatsu Y. 2014. Determination of radioactive cesium isotope ratios by triple quadrupole ICP-MS and its application to rainwater following the Fukushima Daiichi Nuclear Power Plant accident. J Anal At Spectrom, 29: 347–351

Oikawa S, Watabe T, Takata H, et al. 2014. Plutonium isotopes and ^{241}Am in surface sediments off the coast of the Japanese islands before and soon after the Fukushima Dai-ichi nuclear power plant accident. J Radioanal Nucl Chem, 188: 1–6

Orr B, Schöppner M, Tinker R, et al. 2013. Detection of radioxenon in Darwin, Australia following the Fukushima Dai-ichi nuclear power plant accident. J Environ Radioact, 126: 40–44

Periáñez R, Suh K S, Byung Il M, et al. 2013. Numerical modeling of the releases of ^{90}Sr from Fukushima to the Ocean: An evaluation of the source term. Environ Sci Technol, 47: 12305–12313

Povinec P, Aoyama M, Biddulph D, et al. 2013a. Cesium, iodine and tritium in NW Pacific waters-a comparison of the Fukushima impact with global fallout. Biogeosciences, 10: 6377–6416

Povinec P, Gera M, Holý K, et al. 2013b. Dispersion of Fukushima radionuclides in the global atmosphere and the ocean. Appl Radiat Isot, 81: 383–392

Povinec P P, Hirose K, Aoyama M. 2012. Radiostrontium in the western North Pacific: Characteristics, behavior, and the Fukushima impact. Environ Sci Technol, 46: 10356–10363

Povinec P P, Hirose K, Aoyama M. 2013c. Fukushima Accident: Radioactivity Impact on the Environment. New York: Elsevier. 382

Rypina I, Jayne S, Yoshida S, et al. 2013. Short-term dispersal of Fukushima-derived radionuclides off Japan: Modeling efforts and model-data intercomparison. Biogeosciences, 10: 1517–1550

Saegusa J, Kikuta Y, Akino H. 2013. Observation of gamma-rays from fallout collected at Ibaraki, Japan, during the Fukushima nuclear accident. Appl Radiat Isot, 77: 56–60

Saito K, Tanihata I, Fujiwara M, et al. 2014. Detailed deposition density maps constructed by large-scale soil sampling for gamma-ray emitting radioactive nuclides from the Fukushima Dai-ichi Nuclear Power Plant accident. J Environ Radioact, 139: 308–319

Sakaguchi A, Steier P, Takahashi Y, et al. 2014. Isotopic compositions of ^{236}U and Pu isotopes in “Black Substances” collected from roadsides in Fukushima Prefecture: Fallout from the Fukushima Dai-ichi Nuclear Power Plant accident. Environ Sci Technol, 48: 3691–3697

Saunier O, Mathieu A, Didier D, et al. 2013. An inverse modeling method to assess the source term of the Fukushima Nuclear Power Plant accident using gamma dose rate observations. Atmos Chem Phys, 13: 11403–11421

Schneider S, Walther C, Bister S, et al. 2013. Plutonium release from Fukushima Daiichi fosters the need for more detailed investigations. Sci Rep, 3: 2988–2993

Schwantes J M, Orton C R, Clark R A. 2012. Analysis of a nuclear accident: fission and activation product releases from the Fukushima Daiichi Nuclear Facility as remote indicators of source identification, extent of release, and state of damaged spent nuclear fuel. Environ Sci Technol, 46: 8621–8627

Shinonaga T, Steier P, Lagos M, et al. 2014. Airborne plutonium and non-natural uranium from the Fukushima DNPP found at 120 km distance a few days after reactor hydrogen explosions. Environ Sci Technol, 48: 3808–3814

Shozugawa K, Nogawa N, Matsuo M. 2012. Deposition of fission and activation products after the Fukushima Dai-ichi nuclear power plant accident. Environ Pollut, 163: 243–247

Steinhauser G. 2014. Fukushima's forgotten radionuclides: A review of the understudied radioactive emissions. Environ Sci Technol, 48: 4649–4663

Steinhauser G, Brandl A, Johnson T E. 2014. Comparison of the Chernobyl and Fukushima nuclear accidents: A review of the environmental impacts. Sci Total Environ, 470: 800–817

Steinhauser G, Schauer V, Shozugawa K. 2013. Concentration of strontium-90 at selected hot spots in Japan. PloS one, 8: e57760

Stohl A, Seibert P, Wotawa G. 2012a. The total release of xenon-133 from the Fukushima Dai-ichi nuclear power plant accident. J Environ Radioact, 112: 155–159

Stohl A, Seibert P, Wotawa G, et al. 2012b. Xenon-133 and caesium-137 releases into the atmosphere from the Fukushima Dai-ichi nuclear power plant: determination of the source term, atmospheric dispersion, and deposition. Atmos Chem Phys , 12: 2313–2343

Sylvester P, Milner T, Jensen J. 2013. Radioactive liquid waste treatment at Fukushima Daiichi. J Chem Technol Biot, 88: 1592–1596

Tagami K, Uchida S, Ishii N, et al. 2013. Estimation of Te-132 distribution in Fukushima prefecture at the early stage of the Fukushima daiichi nuclear power plant reactor failures. Environ Sci Technol, 47: 5007–5012

Tagami K, Uchida S, Uchihori Y, et al. 2011. Specific activity and activity ratios of radionuclides in soil collected about 20km from the Fukushima Daiichi Nuclear Power Plant: Radionuclide release to the south and southwest. Sci Total Environ, 409: 4885–4888

Tanabe F. 2012. Analyses of core melt and re-melt in the Fukushima Daiichi nuclear reactors: Fukushima NPP Accident Related. J Nucl Sci Technol, 49: 18–36

Tanaka K, Shimada A, Hoshi A, et al. 2014. Radiochemical analysis of rubble and trees collected from Fukushima Daiichi Nuclear Power Station. J Nucl Sci Technol, 51: 1–12

Ten Hoeve J E, Jacobson M Z. 2012. Worldwide health effects of the Fukushima Daiichi nuclear accident. Energy Environ Sci, 5: 8743–8757

TEPCO. 2011. Influence to surrounding environment. http://www.tepco.co.jp/en/nu/fukushima-np/f1/index2-e.html.

Thakur P, Ballard S, Nelson R. 2013. An overview of Fukushima radionuclides measured in the northern hemisphere. Sci Total Environ, 458: 577–613

Tsumune D, Tsubono T, Aoyama M, et al. 2012. Distribution of oceanic ^{137}Cs from the Fukushima Dai-ichi Nuclear Power Plant simulated numerically by a regional ocean model. J Environ Radioact, 111: 100–108

Tsumune D, Tsubono T, Aoyama M, et al. 2013. One-year, regional-scale simulation of ^{137}Cs radioactivity in the ocean following the Fukushima Daiichi Nuclear Power Plant accident. Biogeosciences, 10: 6259–6314

Tumey S, Guilderson T, Brown T, et al. 2013. Input of ^{129}I into the western Pacific Ocean resulting from the Fukushima nuclear event. J Radioanal Nucl Chem, 296: 957–962

UNSEAR. 2008. Effects of Ionizing Radiation: Report to the General Assembly. New York: United Nations Publication. 313

Winiarek V, Bocquet M, Saunier O, et al. 2012. Estimation of errors in the inverse modeling of accidental release of atmospheric pollutant: Application to the reconstruction of the cesium-137 and iodine-131 source terms from the Fukushima Daiichi power plant. J Geophys Res, 117, doi: 10.1029/2011JD016932

Yamamoto M, Sakaguchi A, Ochiai S, et al. 2014a. Isotopic compositions of transuranic nuclides released by the Fukushima Dai-ichi Nuclear Power Plant accident: With emphasis on Cm isotopes. J Radioanal Nucl Chem, 300: 1045–1052

Yamamoto M, Sakaguchi A, Ochiai S, et al. 2014b. Isotopic Pu, Am and Cm signatures in environmental samples contaminated by the Fukushima Dai-ichi Nuclear Power Plant accident. J Environ Radioact, 132: 31–46

Yamamoto M, Takada T, Nagao S, et al. 2012. An early survey of the radioactive contamination of soil due to the Fukushima Dai-ichi Nuclear Power Plant accident, with emphasis on plutonium analysis. Geochem J, 46: 341–353

Yamamoto T. 2012. Radioactivity of fission product and heavy nuclides deposited on soil in Fukushima Dai-Ichi Nuclear Power Plant accident: Fukushima NPP Accident Related. J Nucl Sci Technol, 49: 1116–1133

Yasunari T J, Stohl A, Hayano R S, et al. 2011. Cesium-137 deposition and contamination of Japanese soils due to the Fukushima nuclear accident. Proc Natl Acad Sci USA, 108: 19530–19534

Yoshida N, Kanda J. 2012. Tracking the Fukushima radionuclides. Science, 336: 1115–1116

ZAMG (Central Institue for Meteorology and Geodynamics). 2011. Accident in the Japanese NPP Fukushima: Large emissions of Cesium-137 and iondine-131. http://www.zamg.ac.at/docs/aktuell/japan2011-03–24_1600_E.pdf

Zheng J, Tagami K, Bu W, et al. 2014. $^{135}Cs/^{137}Cs$ isotopic ratio as a new tracer of radiocesium released from the Fukushima nuclear accident. Environ Sci Technol, 48: 5433–5438

Zheng J, Tagami K, Uchida S. 2013. Release of plutonium isotopes into the environment from the Fukushima Daiichi nuclear power plant accident: What is known and what needs to be known. Environ Sci Technol, 47: 9584–9595

Zheng J, Tagami K, Watanabe Y, et al. 2012. Isotopic evidence of plutonium release into the environment from the Fukushima DNPP accident. Sci Rep, 2: 304–312