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Importance of Accounting for the Partitioning of Iodine Released During Nuclear Power Plant Accidents

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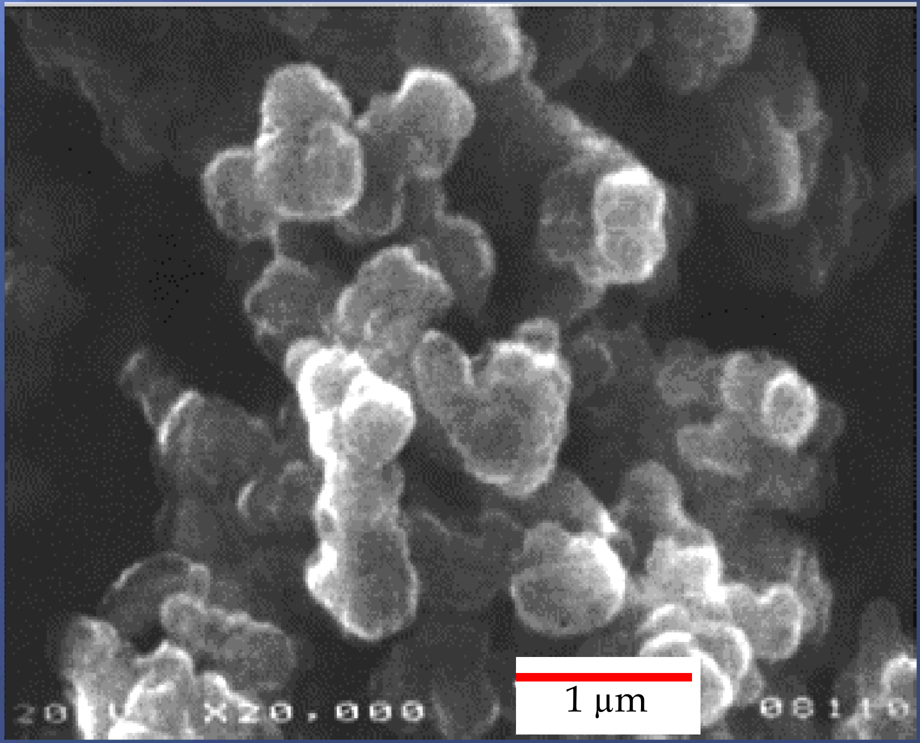
Why Do We Care About Iodine?

What is Iodine Partitioning?

- Iodine released from a nuclear power plant (NPP) can exist in multiple chemical and physical forms.
- Gaseous Iodine:
  - Organic or non-reactive (e.g., CH_3I)
  - Inorganic or reactive, (e.g., I_2)
- Particulate Iodine:
  - Varying particle size distributions
  - Big, ~ 1 - 5 µm
  - Small particle species (e.g., CsI, AgI), ~ 0.1 – 1.0 µm
  - Very small particles species (e.g., I_xO_y), ~ 0.02 µm



Why Do We Care About Iodine Partitioning?

- Inhalation dose coefficients (DCs) vary by:
  - Physical and chemical form
  - Particle size distribution (PSD)
- Deposition velocity is function of chemical and physical form
- Therefore, we need to know how iodine is partitioned in order to:
  - Perform atmospheric dispersion modeling to predict downwind air and ground concentrations,
  - Perform accurate radiological dose assessments /dose projections and
  - Provide suitable information to help decision makers make appropriate protective action decisions (e.g., sheltering, evacuation, food interdiction).

Inhalation Dose Coefficients for Different Forms of Iodine

Organ	I-131 ICRP 60 Series Inhalation Dose Coefficients						Ratio		Ratio	
	Particulate (1 µm AMAD, Class F)		Gas/Vapor: Reactive (I_2)		Gas/Vapor: Non-Reactive (CH_3I)		I_2/Particulate		CH_3I/Particulate	
	Adult	1-Year Old	Adult	1-Year Old	Adult	1-Year Old				
	(Sv/Bq)	(Sv/Bq)	(Sv/Bq)	(Sv/Bq)	(Sv/Bq)	(Sv/Bq)	Adult	1-Year Old	Adult	1-Year Old
Thyroid	1.46E-07	1.42E-06	3.93E-07	3.24E-06	3.07E-07	2.53E-06	2.67	2.27	2.09	1.77
Effective	7.34E-09	7.12E-08	1.98E-08	1.63E-07	1.54E-08	1.27E-07	2.68	2.27	2.08	1.77

- Inhalation DCs for reactive and non-reactive forms of iodine are significantly (~1.7 to 2.7) greater than that for particulate form.
- Inhalation DCs for reactive form of iodine are ~28% larger than those for the non-reactive forms of iodine

Iodine Inhalation Dose Coefficients for Different PSDs

Organ	I-131, Class F, ICRP 60 Series Inhalation Dose Coefficients (DCFPAK 2.2)									
	Particulate (0.5 µm AMAD)		Particulate (1 µm AMAD)		Particulate (2 µm AMAD)		Particulate (5 µm AMAD)		Particulate (8 µm AMAD)	
	Adult	1-Year Old	Adult	1-Year Old	Adult	1-Year Old	Adult	1-Year Old	Adult	1-Year Old
	(Sv/Bq)	(Sv/Bq)	(Sv/Bq)	(Sv/Bq)	(Sv/Bq)	(Sv/Bq)	(Sv/Bq)	(Sv/Bq)	(Sv/Bq)	(Sv/Bq)
Thyroid	1.09E-7	1.16E-6	1.46E-07	1.42E-06	1.91E-7	1.68E-6	2.10E-7	1.70E-6	1.97E-7	1.57E-6
Effective	5.50E-9	5.82E-8	7.34E-09	7.12E-08	9.59E-9	8.42E-8	1.06E-8	8.54E-8	9.93E-9	7.89E-8

For the PSDs shown, the largest Thyroid and Effective DCs are associated with 5 µm AMAD PSD for both Adult and 1-year old age groups.

What Iodine Partitioning is Appropriate?

- Inhalation DCs vary with the form of iodine and with the particle size of the aerosol fraction.
- Therefore,
  - assuming all iodine is particulate or gaseous leads to inaccurate dose projections and
  - assuming all particulate iodine has one PSD can lead to inaccurate dose projections.

- What is the appropriate partitioning of iodine for a NPP accident?
- What is the appropriate iodine PSD for NPP accident?

Modeling Iodine from NPP

- We know a lot about the physical forms of iodine inside NPP containment from various studies (e.g., PHÉBUS – FP experiments).
- Physical forms of iodine (e.g., particulate, reactive gas, non-reactive gas) in containment are not necessarily the forms of iodine that are transported down wind and deposited on the ground.
- The ratio of the physical forms released from containment are not fixed.
- Physical forms released to the atmosphere are converted in the environment (e.g., photochemistry).
- Physical forms of iodine may vary with the distance from the release point (e.g., particulates fall out quicker than gaseous forms).

Iodine Partitioning Report in Support of HEDR

- Primary objective of the Hanford Environmental Dose Reconstruction (HEDR) Project is to develop individual and population dose estimates from operations conducted at Hanford since 1944.
- Reviews studies of partitioning of iodine in stacks and the environment.
- Iodine exists in the environment as particulate, reactive (inorganic) gas and non-reactive (organic) gas.
- Near the release point, the 1963 experiments of reactive iodine (I_2)"…**clearly show the particulate and non-reactive iodine fractions increasing with distance and the reactive fraction decreasing with distance."**
- "Weapons test data do not appear to be a good source of information; the iodine partitioning in fallout debris is distinctly different the from the partition found from other sources."

Reference: "Partitioning Iodine for Deposition Calculations in the Atmospheric Transport Model," Draft Report, Hanford Environmental Dose Reconstruction Project , J.V. Ramsdell, Pacific Northwest Laboratories, Richland, WA, July 1992

Iodine Partitioning Implemented by RASCAL 4

- NRC's RASCAL 4 software code implements iodine partitioning assumptions based upon data from the Hanford Environmental Dose Reconstruction (HEDR) Project, Chernobyl and atmospheric iodine reports (See NUREG-1940).
- Assumes partitioning reaches an equilibrium within a few kilometers of the release point and remains constant
- Iodine partitioning:
  - 25% is particulate
  - 30% is I_2 (reactive gas) and
  - 45% is CH_3I (non-reactive gas)
- Assumes particulate fraction is 1 µm and uses lung clearance class with highest dose coefficient

Iodine Partitioning Observed by EPA

EPA's RadNet Monitoring data collected during Fukushima Response

Data provided by Lowell Ralston (U.S. EPA)

Location	Collected	Charcoal/Glass-2"	2σ uncert.	Charcoal/Glass-4"	2σ uncert.
DUTCH HARBOR	3/19/2011	3.6	0.7	3.5	0.6
	3/20/2011	---	---	13.7	3.0
	3/22/2011	10.1	5.0	9.9	2.4
	3/23/2011	---	---	1.9	2.2
JUNEAU	3/22/2011	2.8	1.2	3.1	1.0
	3/23/2011	3.2	1.3	3.8	1.0
	3/24/2011	6.3	4.0	---	---
NOME	3/21/2011	6.0	2.5	---	---
	3/22/2011	5.0	2.1	3.5	0.8
	3/23/2011	3.6	0.8	4.0	0.7
	3/24/2011	3.4	0.7	3.8	0.7
NAREL	3/24/2011	4.8	1.9	---	---
ANAHEIM	3/20/2011	6.8	1.5	6.9	1.2
	3/21/2011	11.4	2.6	12.5	2.2
	3/22/2011	5.7	1.7	6.6	1.4
SAN BERN. CTY.	3/20/2011	4.8	1.0	4.2	0.7
	3/22/2011	9.6	2.8	9.5	1.8
	3/23/2011	4.8	1.6	4.8	1.1
SAIPAN	3/21/2011	2.9	0.9	3.3	0.8
	3/22/2011	6.1	2.4	6.7	1.6
GUAM	3/21/2011	---		4.9	1.1

Note: Charcoal (iodine gas) and Glass (particulate iodine)

Iodine Partitioning Observed by EPA

EPA's RadNet Monitoring data collected during Fukushima Response

Data provided by Lowell Ralston (U.S. EPA)

Location	Collected	Charcoal/Glass-2"	2σ uncert.	Charcoal/Glass-4"	2σ uncert.
KAHUKU	3/20/2011	3.7	0.9	5.3	1.1
	3/21/2011	2.1	0.4	2.0	0.4
	3/23/2011	13.7	5.6	15.1	3.3
	3/23/2011	5.8	1.8	7.2	1.6
KAUAI	3/21/2011	2.1	0.4	2.8	0.5
	3/22/2011	2.1	1.0	2.6	0.9
BOISE	3/21/2011	3.8	1.2	4.5	0.9
	3/22/2011	---	---	6.7	1.4
	3/23/2011	5.6	4.0	8.9	3.0
LAS VEGAS	3/18/2011	1.9	1.0	---	---
	3/21/2011	5.0	1.0	---	---
	3/22/2011	10.8	5.0	---	---
	3/23/2011	6.5	3.6	---	---
	3/24/2011	3.1	0.8	---	---

	Charcoal/Glass-2"	Charcoal/Glass-4"
Count:	31	27
Mean:	5	6
Median	5	5
Std, Dev:	3	4
Min:	2	2
Max:	14	15

Note: Charcoal (iodine gas) and Glass (particulate iodine)

Summary of EPA Iodine Partitioning Data

- On average, ~ 5-6 times more iodine radioactivity was in gaseous forms compared to particulate forms of iodine.
- Reactive and non-reactive gaseous forms were not distinguished.

NOTE: All EPA data collected a long way from release point

EPA's RadNet monitoring stations

Iodine Partitioning Observed by IMS During Fukushima NPP Accident Response

- International Monitoring System (IMS) is a global monitoring system established to verify compliance with the Comprehensive Nuclear-Test-Ban Treaty.
- Includes 80 stations across the Earth capable of measuring particulate radionuclides (high-volume, 900 – 1000 m^3/hr, 24 hr samples).
- 40 of the stations capable of monitoring particulates and noble gases.
- Although the IMS radionuclide network is not required to use activated charcoal traps (i.e., incapable of measuring gaseous iodine), the IMS network had access to data from the “Ring of Five (Ro5)” network in Europe that provided particulate and gaseous iodine data.

Iodine Partitioning Observed by IMS During Fukushima NPP Accident Response

- "Based on the results provided by Ro5:
  - the average gaseous/total ratio for I-131 is 77.2% +/-
  - 13.6% (Masson et al., (2011)). This value is close to the average reported after the Chernobyl accident (Cambray et al., 1987) and to the 71% +/- 11% average reported from Fukushima NPP site during the period from 22 March to 4 April.
  - **Therefore, 4 times more iodine activity observed in gaseous phase compared to the particulate phase.**

NOTE: All Ro5 data collected a long way from release point

Reference: "Detection of Radionuclides emitted During the Fukushima Nuclear Accident with the CTBT Radionuclide Network," 2011 Monitoring Research Review: Ground-Based Nuclear Explosion Monitoring Technologies, Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization, U.Stoehlker, M. Nikkinen and A. Gheddou

Iodine Partitioning Observed by DOE/NNSA During Fukushima NPP Accident Response

- NNSA/DOE conducted air monitoring while deployed to Japan in support of Operation Tomodachi
- Air monitors:
  - had ability to collect particulate and gaseous forms of iodine and
  - did not have ability to separate inorganic (e.g., I_2) and organic (CH_3I) forms of iodine gas
- Tabulated data on next slide:
  - Does not include entire NNSA/DOE dataset,
  - Data was sorted by activity
  - Data only includes results > 1E-10 µCi/m^3 (particulate)

Iodine Partitioning Observed by CM During Fukushima NPP Accident Response

Particulate Sample Number	Gas Sample Number	Location	Mid-Point Collection Time	Radio-Nuclide	Reported Paper Filter Sample Activity (µCi/ml)	Reported Charcoal Filter Sample Activity (µCi/ml)	Total Reported Sample Activity (µCi/ml)	Fraction of Total I-131 Activity Due to Particulate I-131	Fraction of Total I-131 Activity Due to Gasous I-131
SCF-00092	SCF-00093	Expressway Exit 17	3/21/2011 13:51	I-131	6.08E-08	2.08E-08	8.16E-08	0.745	0.255
SCF-00094	SCF-00095	?	?	I-131	1.71E-08	5.87E-09	2.29E-08	0.747	0.256
SCF-00086	SCF-00087	Oarai	3/22/2011 15:48	I-131	5.86E-09	1.92E-09	7.78E-09	0.753	0.247
SCF-00139	SCF-00140	6 Km from NPP	3/23/2011 14:53	I-131	4.36E-09	3.70E-09	8.06E-09	0.541	0.459
SCF-00006	SCF-00007	Outside Narita	3/22/2011 13:50	I-131	1.93E-09	5.49E-10	2.48E-09	0.778	0.221
SCF-00088	SCF-00089	Mito	3/22/2011 16:32	I-131	1.84E-09	9.02E-10	2.74E-09	0.672	0.329
SCF-00008	SCF-00085	Ibaraki	3/22/2011 14:44	I-131	1.35E-09	4.85E-10	1.84E-09	0.734	0.264
SCF-00090	SCF-00091	Expressway Exit 18	3/21/2011 13:17	I-131	1.35E-09	1.17E-09	2.52E-09	0.536	0.464
SCF-08991	SCF-00516	Exit at Fukishima	3/20/2011 17:39	I-131	1.33E-09	8.75E-12	1.34E-09	0.993	0.007
SCF-00041	SCF-00042	Aerial over Fukushi	3/20/2011 13:48	I-131	1.05E-09	6.94E-10	1.74E-09	0.603	0.399
SCF-00083	SCF-00084	Aerial, Hello	3/22/2011 16:54	I-131	1.05E-09	8.31E-10	1.88E-09	0.559	0.442
SCF-00004	SCF-00005	Chiba Dock	3/22/2011 12:03	I-131	1.01E-09	3.86E-10	1.40E-09	0.721	0.276
SCF-00142	SCF-00143	Road	3/23/2011 15:53	I-131	9.44E-10	5.27E-10	1.47E-09	0.642	0.359
SCF-00151	SCF-00152	Expressway Exit Hit	3/23/2011 14:19	I-131	6.10E-10	2.08E-10	8.18E-10	0.746	0.254
SCF-00235	SCF-00236	Fukushima Valley	3/24/2011 14:11	I-131	4.23E-10	1.95E-10	6.18E-10	0.684	0.316
SCF-07671	SCF-07672	20 mi S of NPP	3/19/2011 13:12	I-131	2.32E-10	2.09E-10	4.41E-10	0.526	0.474
SCF-00124	SCF-00125	Embassy	3/23/2011 0:57	I-131	1.97E-10	3.10E-10	5.07E-10	0.389	0.611
SCF-00096	SCF-00097	Expressway Exit 15	3/21/2011 14:53	I-131	1.72E-10	3.09E-09	3.27E-09	0.053	0.945
SCF-00107	SCF-00108	Bldg. 1503	3/21/2011 14:13	I-131	1.39E-10	5.03E-10	6.41E-10	0.217	0.785
SCF-00035	SCF-00036	Harris Tower	3/21/2011 1:15	I-131	1.30E-10	2.06E-10	3.36E-10	0.387	0.613
SCF-00132	SCF-00133	Bldg. 1503	3/22/2011 17:10	I-131	1.19E-10	1.04E-10	2.24E-10	0.531	0.464
							Number =	21	21
							Mean =	0.598	0.402
							Median =	0.642	0.359
							Std. Dev. =	0.209	0.209
							Minimum =	0.053	0.007
							Maximum =	0.993	0.945

MELCOR Predicted Iodine Particle Size Distribution - BWR

Predicted PSD for particulate iodine based on the Peach Bottom (BWR) Long-Term Station Blackout Uncertainty Analysis

- "...predominate particle size bin is ~0.53 µm."
- "... a good estimate could be considered between the ranges of 0.3 – 1.0 µm based on the data."

Particle Size Bin	Mass Median Aerosol Diameter (µm)	BWR Long-term Station Blackout Uncertainty Analysis	
		Mean	Median
Bin 1	0.15	7.01%	5.39%
Bin 2	0.29	25.67%	25.60%
Bin 3	0.53	29.06%	29.10%
Bin 4	0.99	21.80%	21.60%
Bin 5	1.8	11.26%	9.97%
Bin 6	3.4	3.92%	2.44%
Bin 7	6.4	0.99%	0.39%
Bin 8	11.9	0.23%	0.04%
Bin 9	22.1	0.05%	0.00%
Bin 10	41.2	0.02%	0.01%

Reference: State-of-the-Art Reactor Consequence Analyses – Uncertainty Analysis of the Unmitigated Long-Term Station Blackout of the Peach Bottom Atomic Power Station, Doug Osborn, Sandia National Laboratories, DRAFT REPORT, NUREG/CR-7155, SAND2012-10702P.

MELCOR Predicted Iodine Particle Size Distribution - PWR

Predicted PSD for particulate iodine based on the Surry Station Blackout (PWR) Dynamic risk Assessment

- “…predominate particle size bin is ~1.8 µm.”
- “… a good estimate could be considered between the ranges of 0.5 – 1.80 µm based on the data.”

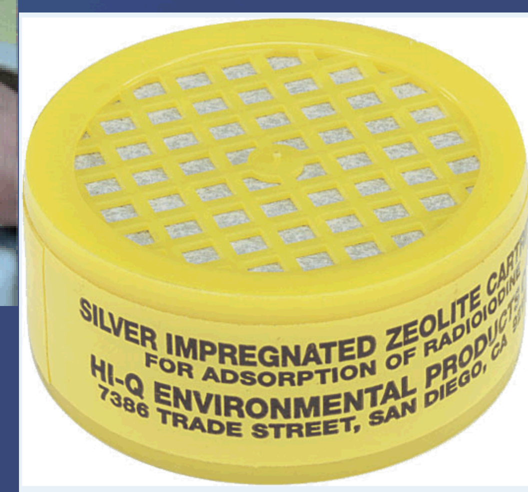
Particle Size Bin	Mass Median Aerosol Diameter (µm)	BWR Long-term Station Blackout Uncertainty Analysis	
		Mean	Median
Bin 1	0.15	0.67%	0.53%
Bin 2	0.29	2.99%	2.74%
Bin 3	0.53	10.51%	10.10%
Bin 4	0.99	26.77%	26.60%
Bin 5	1.8	33.92%	34.90%
Bin 6	3.4	18.78%	19.50%
Bin 7	6.4	4.96%	4.22%
Bin 8	11.9	0.93%	0.37%
Bin 9	22.1	0.29%	0.02%
Bin 10	41.2	0.18%	0.01%

Reference: “Seamless Level 2/Level 3 Probabilistic Risk Assessment using Dynamic Event Tree Analysis,” PhD Dissertation, Doug Osborn, Sandia National Laboratories, 2013 (Draft)

Determining Iodine Partitioning

- Aerosol/Particulate (paper) filter and cartridge (activated carbon)air samplers
- Particulate filter captures most particle sizes with high capture efficiency (very small particles can pass through)





Determining Iodine Partitioning

- Cartridges of activated carbon impregnated with silver zeolite or triethylene diamine (TEDA):
  - capture non-particulate iodine with high efficiently, especially at low flow rates.
  - Advantage of silver zeolite is that it captures and retains much less noble gas (e.g., Xe) than TEDA.
  - Does not distinguish between reactive and non-reactive forms of gaseous iodine. However, placing a silver-wire mesh between the paper and the carbon will enable the 3 forms of iodine to be distinguished. The paper collects the particulates, the silver wire collects the reactive gas and the carbon collects the non-reactive gas.

Determining Iodine PSDs

- Cascade impactors
- Electron microscopes
- Spectrometers
- Others?

- Not really practical to determine iodine PSDs for Early Phase protective action decisions.
- Therefore, must make some assumptions.

Conclusions

- There is no one iodine partitioning assumption that is perfect for iodine releases from all facilities and by all release mechanisms.
- There is no one iodine particle size distribution assumption that is perfect for all iodine releases
- NRC's assumptions implemented in RASCAL 4 seem to be a good starting point for modeling.
- Goal should be to have all modelers implement similar default model input assumptions to ensure consistency between predictions and radiological dose assessments.

Questions?