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Dose Implications of Radioiodine Speciation with a Case Study Using CANDU Severe Accidents

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Outline

1. Background: “Why understanding iodine speciation is important”

2. Source Term Modeling

3. Atmospheric Modeling

Background

Why understanding iodine speciation is important

Radioiodine is the Single Most Important Contributor to Public Doses During Accidents

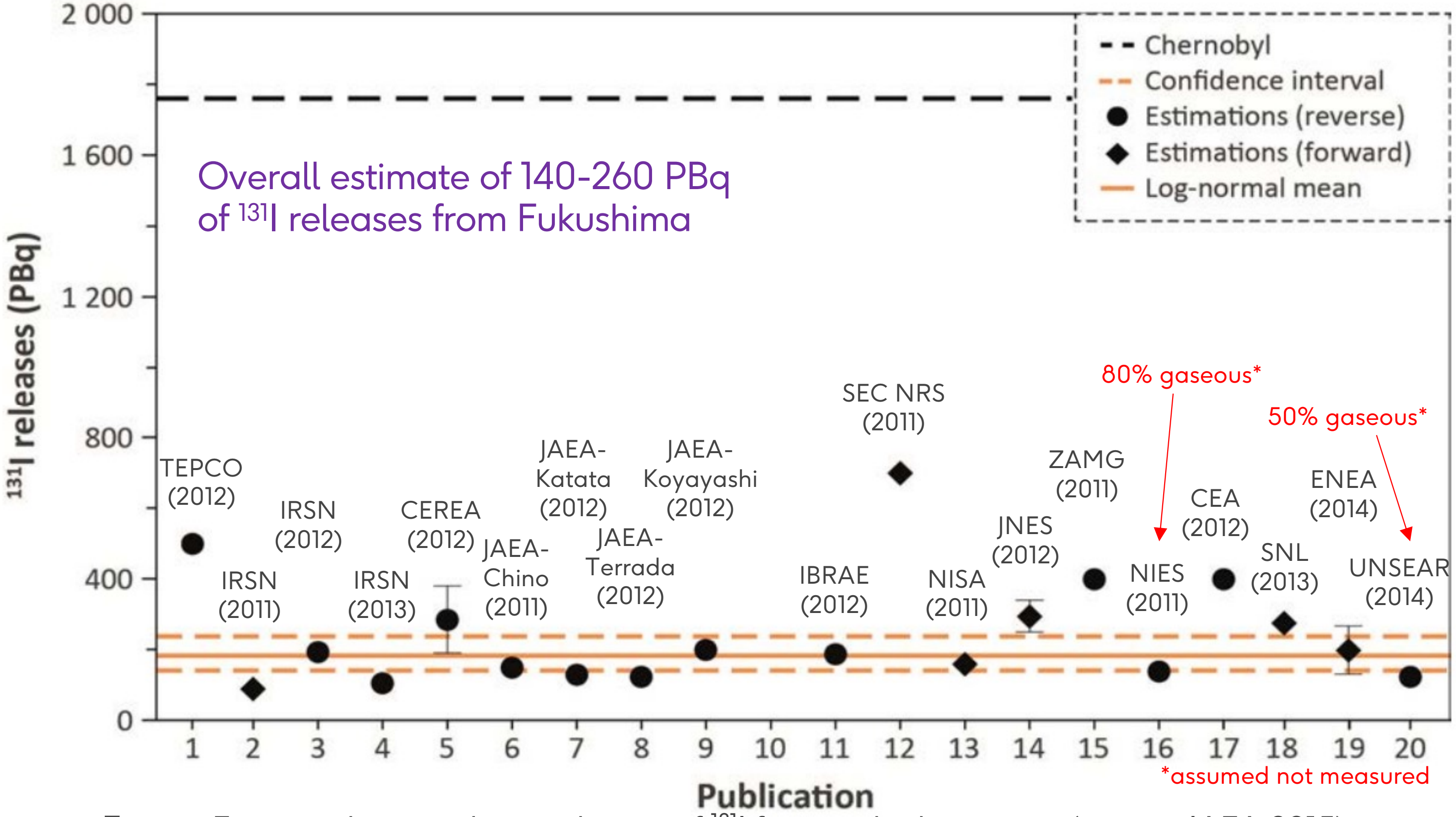
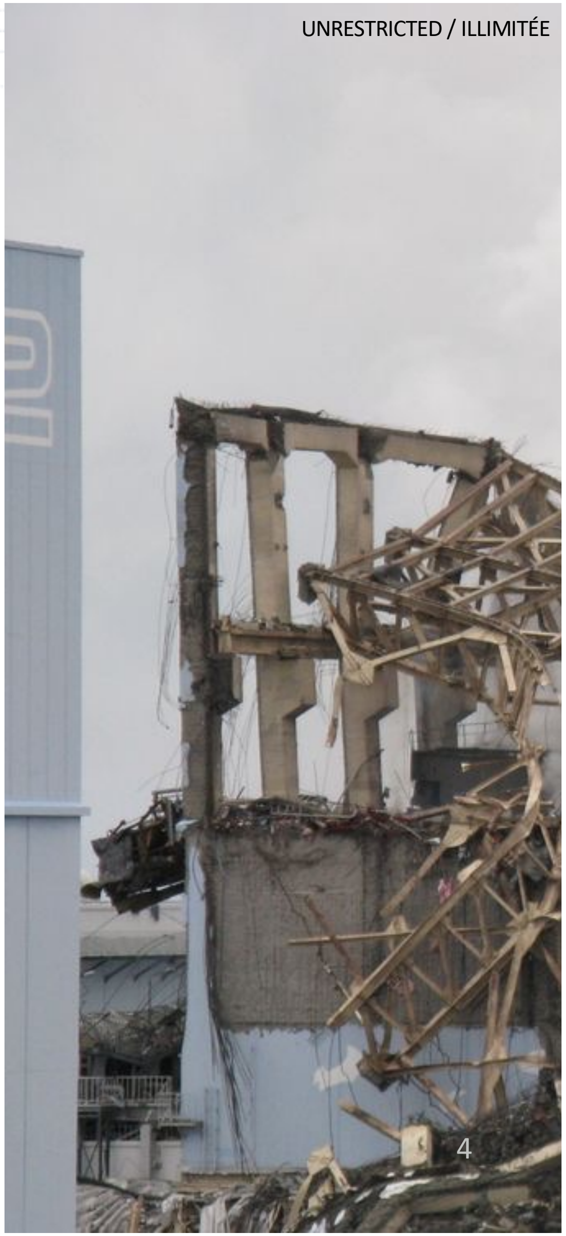


Figure: Estimated atmospheric releases of ^{131}I from multiple sources (source: IAEA 2015).



Source Term Dynamics

Simplified assumptions of speciation:

- NRC NUREG-1465: 95% as CsI, remainder gaseous,
- ICRP 158: 50% aerosol / 50% gaseous.

Realistically, several factors influence source term speciation and the conversion of CsI into elemental and organic iodine vapours, and oxide aerosols:

- Transport and re-volatilization in the reactor cooling system (RCS),
- Aerosol physics in containment,
- Liquid phase chemistry,
- Gas phase chemistry,
- Safety system interactions and timing of failures.

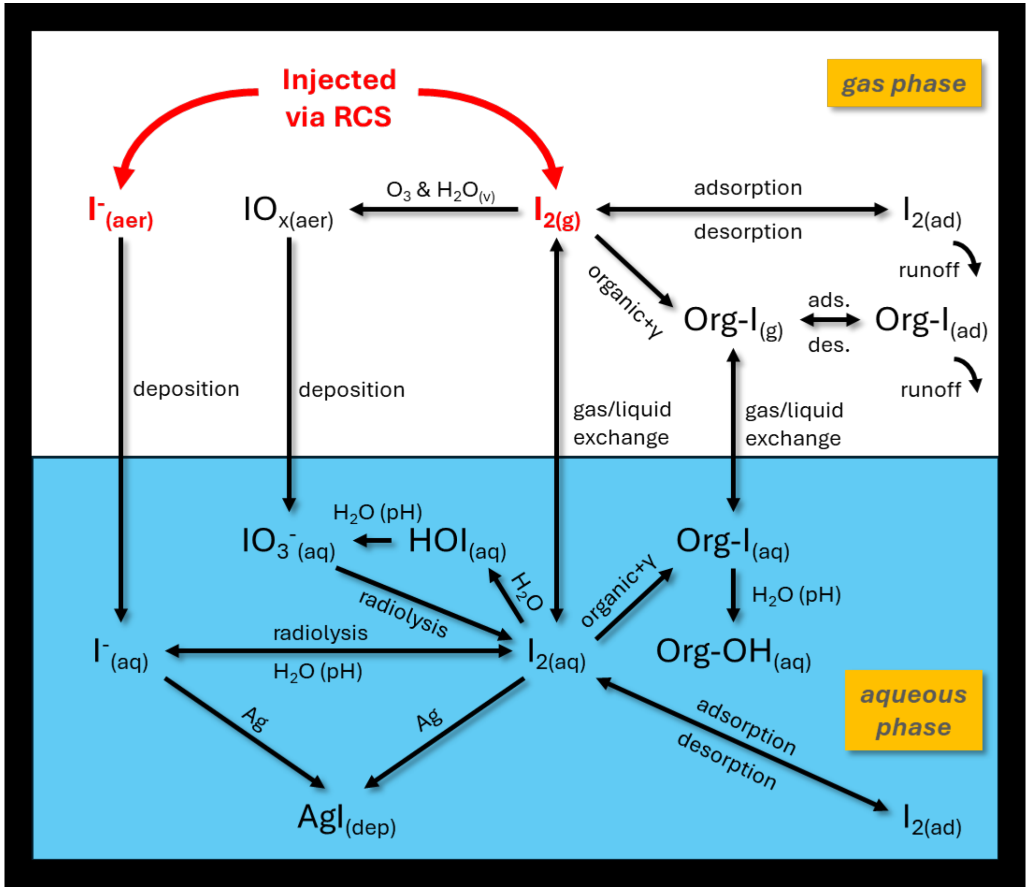


Figure: Simplified diagram of iodine transformations within containment (modified from OECD/NEA 2007).

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Atmospheric Behaviour

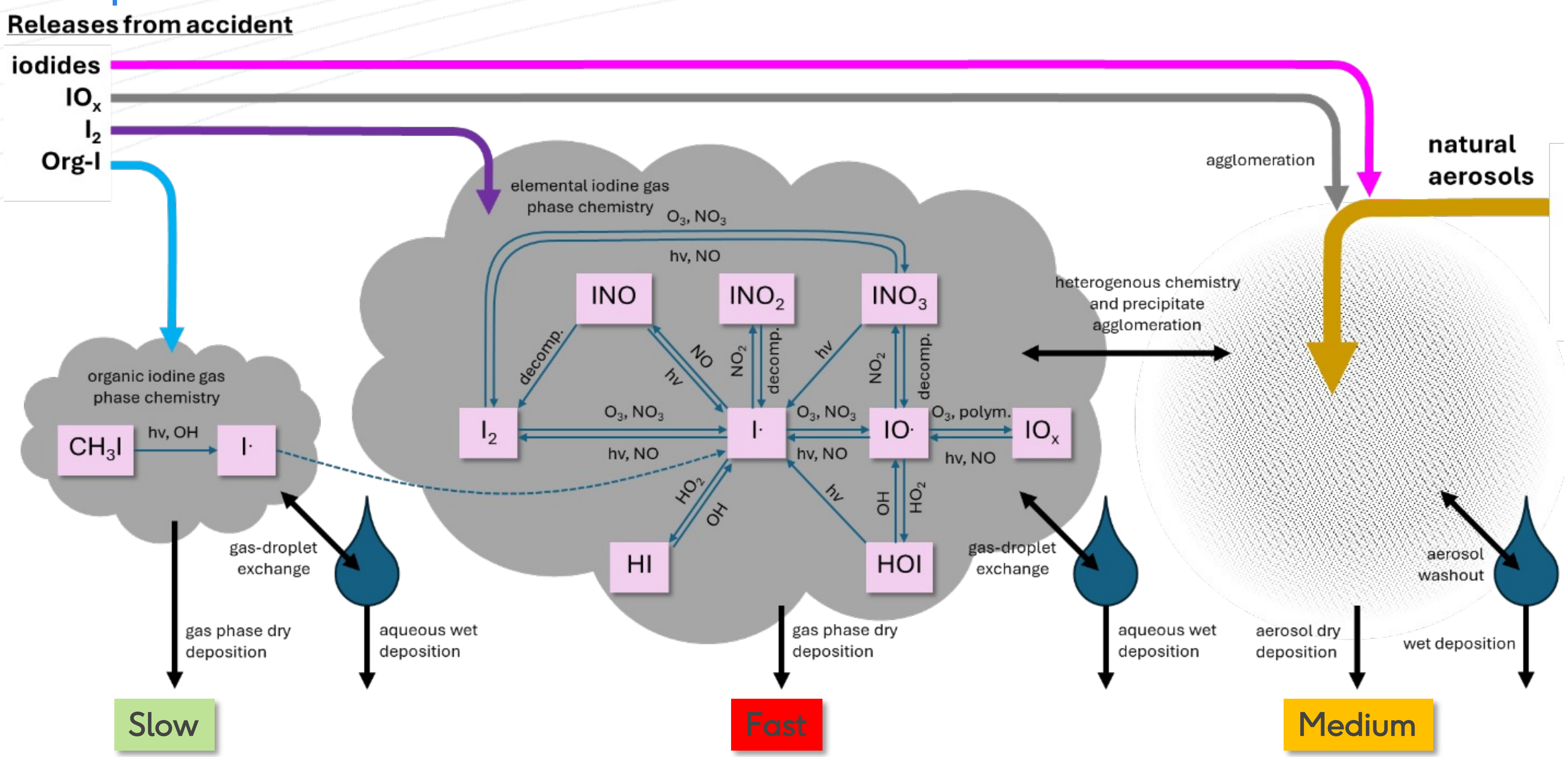


Figure: Further atmospheric physical and chemical interactions that lead to the photolytic destruction of gaseous species, interactions with atmospheric pollutants and natural aerosols, as well as dry and wet deposition.

Fukushima Daiichi Accident Observations

Many studies available on global scale (e.g., Masson et al. 2019 review), but a more limited number from Japan from which close range (<200 km) capable of providing data on detailed physiochemical characteristics.

Publication	Description
Chino et al. (2011)	Sampling of aerosol radionuclides (simple filters), source reconstruction.
Doi et al. (2013)	Sampling of gaseous and aerosol radionuclides, incl. cascade impactor of particle size distribution, different behaviour of iodine and cesium.
Xu et al. (2015)	Re-analysis of Doi (2013) data, comparing proportion of gaseous and aerosol forms.
TEPCO	Sampling of gaseous and aerosol radionuclides (simple filters).
JAEA	Sampling of gaseous and aerosol radionuclides (simple filters).
Lebel et al. (2016)	Re-analysis of TEPCO and JAEA data, inferring speciation from filter penetration and nuclide ratios.
Adachi et al. (2013)	Sampling of aerosol radionuclides, SEM and EDS analysis for inference of insoluble Cs.
Kaneyasu (2012)	Sampling of gaseous and aerosol radionuclides, incl. cascade impactor, inference of Cs particulate interactions with background aerosols.

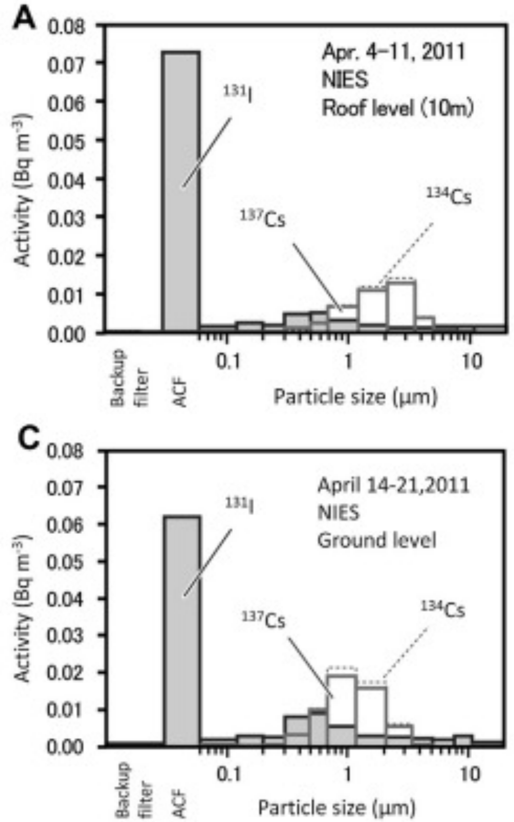


Figure: Particle size distributions and for radionuclide containing aerosols & volatile iodine capture at Tsukuba (source: Doi 2013).

Health Impacts of Radioiodine Species

- Iodine compounds have a high affinity toward different organs, mainly the thyroid.
- **Factor of ~2 difference in dose coefficients for gaseous vs soluble aerosol (type F) forms.**
- ^{132}Te is also an important contributor to iodine dose, due to equilibrium with ^{132}I.
- Inhalation component dominant dose pathway during early phase of accident, with uptake from food more important later.

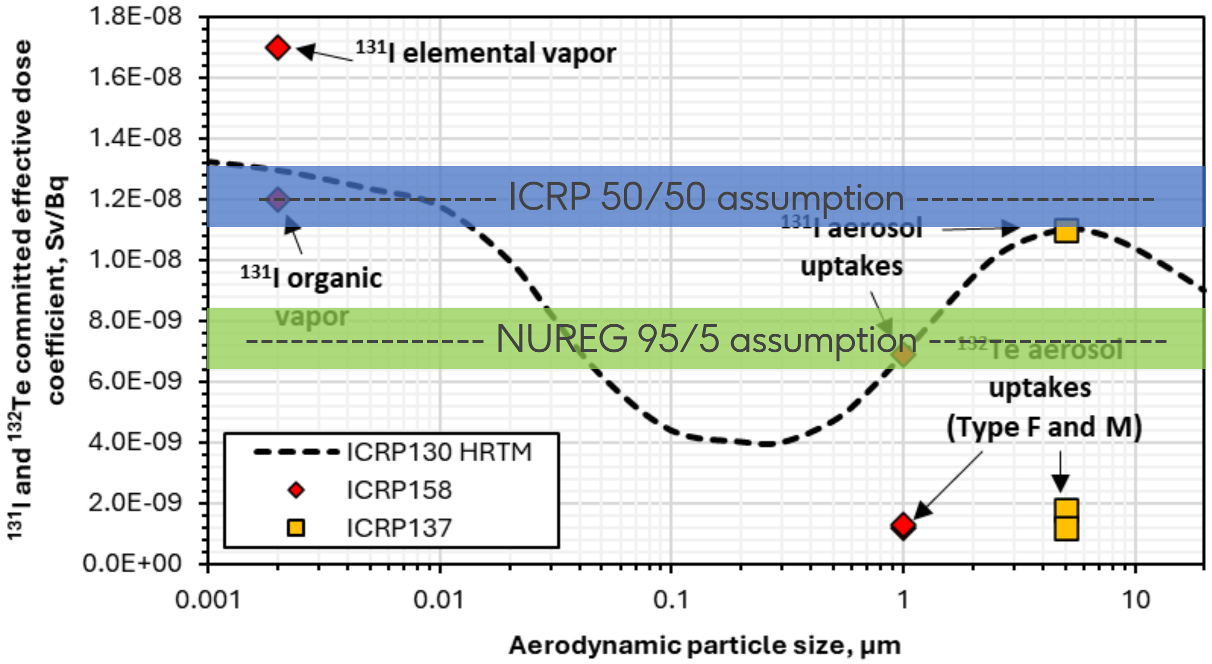


Figure: Committed effective whole body dose coefficients for the inhalation or ingestion of ^{131}I and ^{132}Te, values from ICRP publications 137 and 158.

Source Term Modeling

Coupling MAAP-CANDU and ASTEC

Modeling Framework

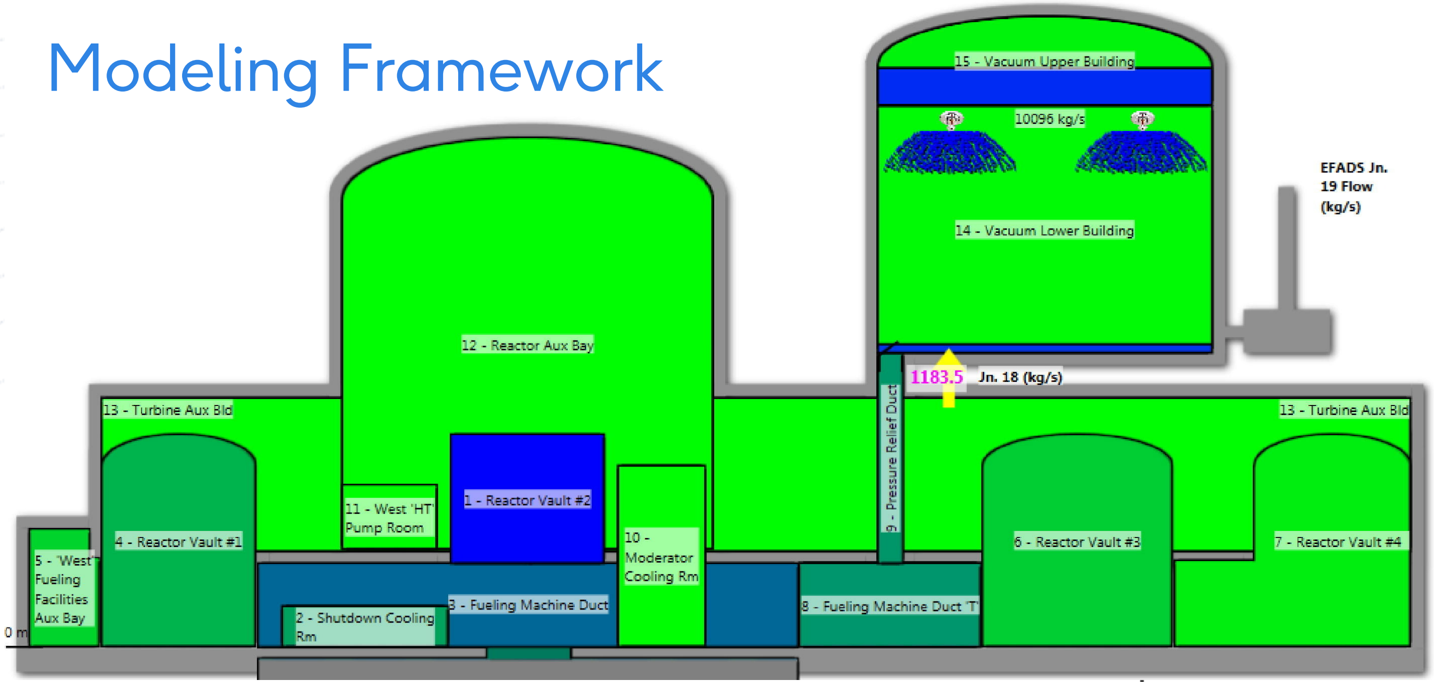
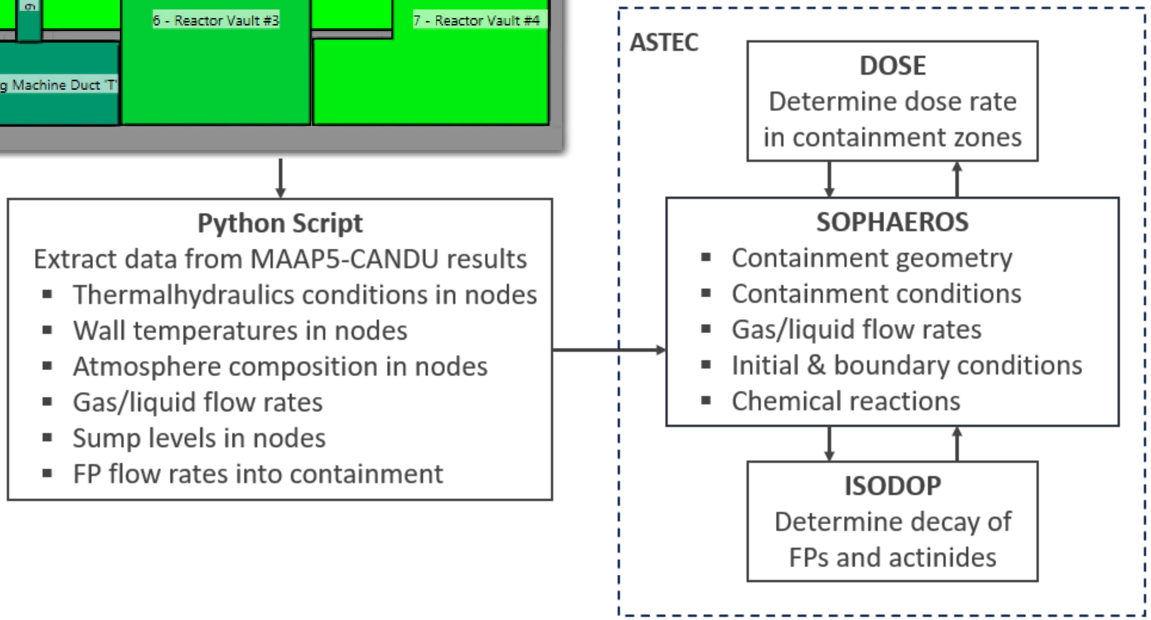


Figure: Workflow of combined MAAP-CANDU (full event sequence including core degradation) and ASTEC calculations (iodine containment chemistry).



Four accident cases considered:

1. Unmitigated SBO with containment failure.
2. SBO mitigated after full core disassembly.
3. SBO with use of only filtered venting.
4. Unmitigated LLOCA with containment failure.

Radioiodine Species Releases

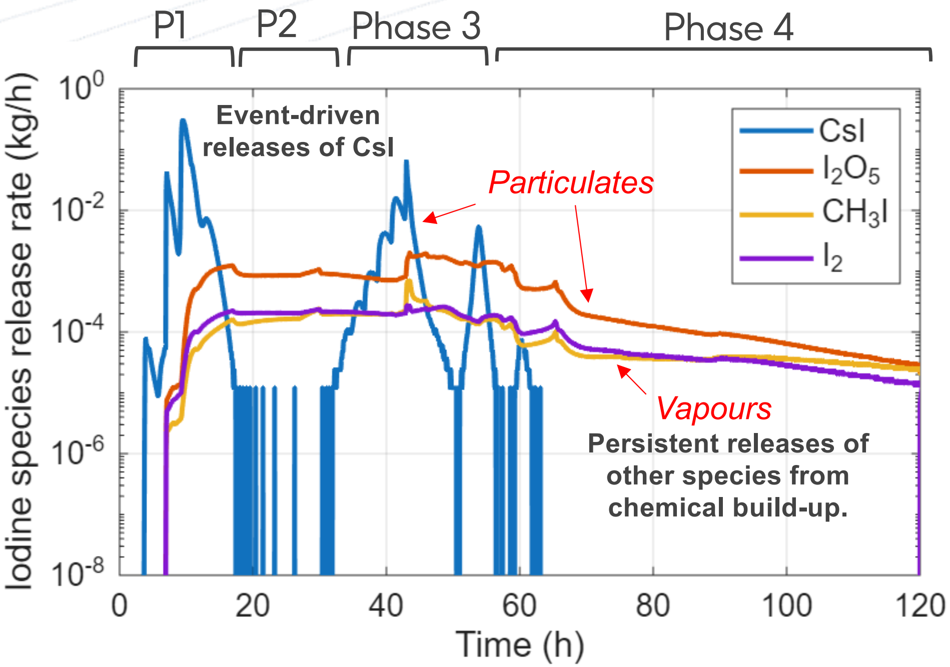


Figure: Case 1 (unmitigated SBO).

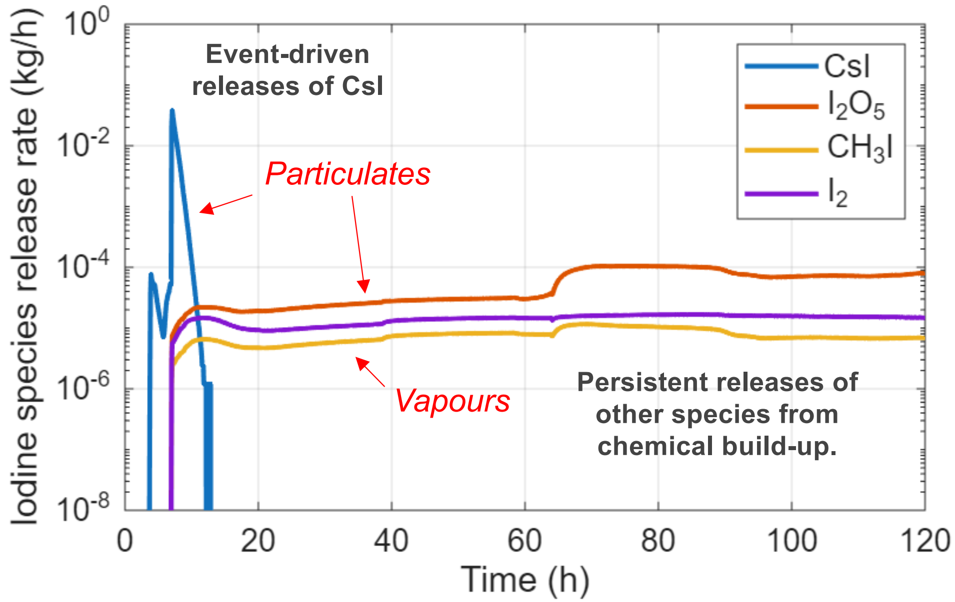


Figure: Case 6 (SBO until end of core disassembly).

CsI releases approx. 0.2 µm
I_2O_5 releases approx. 0.3-1.0 µm

Atmospheric Modeling

F0AM box model of full chemistry suite

Modeling Framework

- **Aim**: To quantify how atmospheric chemistry of released iodine species affects:
  - Chemical speciation during plume transport,
  - Inhalation dose coefficients,
  - Ground deposition behavior.
- **Approach**: 0-D atmospheric box modelling using the F0AM framework.
- **Simulation Runs:** Time-resolved iodine source terms broken into four phases:
  - Separate runs for each accident case (1, 6, 9, 10), release phase (1–4), and day or night,
  - Precursor sims run for 24 h to obtain realistic background O_3 and VOC/NO_x ratios,
  - Then iodine species released and dispersion run for 8 h,
  - Constant meteorology (wind speed of 3 m/s, Pasquill Class D stability).

Geosci. Model Dev., 9, 3309–3319, 2016
www.geosci-model-dev.net/9/3309/2016/
doi:10.5194/gmd-9-3309-2016

Geoscientific Model Development
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The Framework for 0-D Atmospheric Modeling (F0AM) v3.1

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- **Base mechanism:** RACM2 (363 reactions).
- **Added custom mechanisms**:
  - **63 iodine reactions –** Inorganic, organic, heterogeneous, and photolysis reactions,
  - **Dry deposition –** CsI & I_2O_5 aerosols, I_2, CH_3I, inorganic iodine species.

Chemical Speciation Results

- **Aerosol forms (CsI and I_2O_5):** most important species commensurate with source term.
- **Vapour forms (I_2 and CH_3I):** each deplete during the day from photolysis (rapidly or slowly), converting to atomic I radicals:
  - *Fraction of organic species grows at night because it has least deposition.*

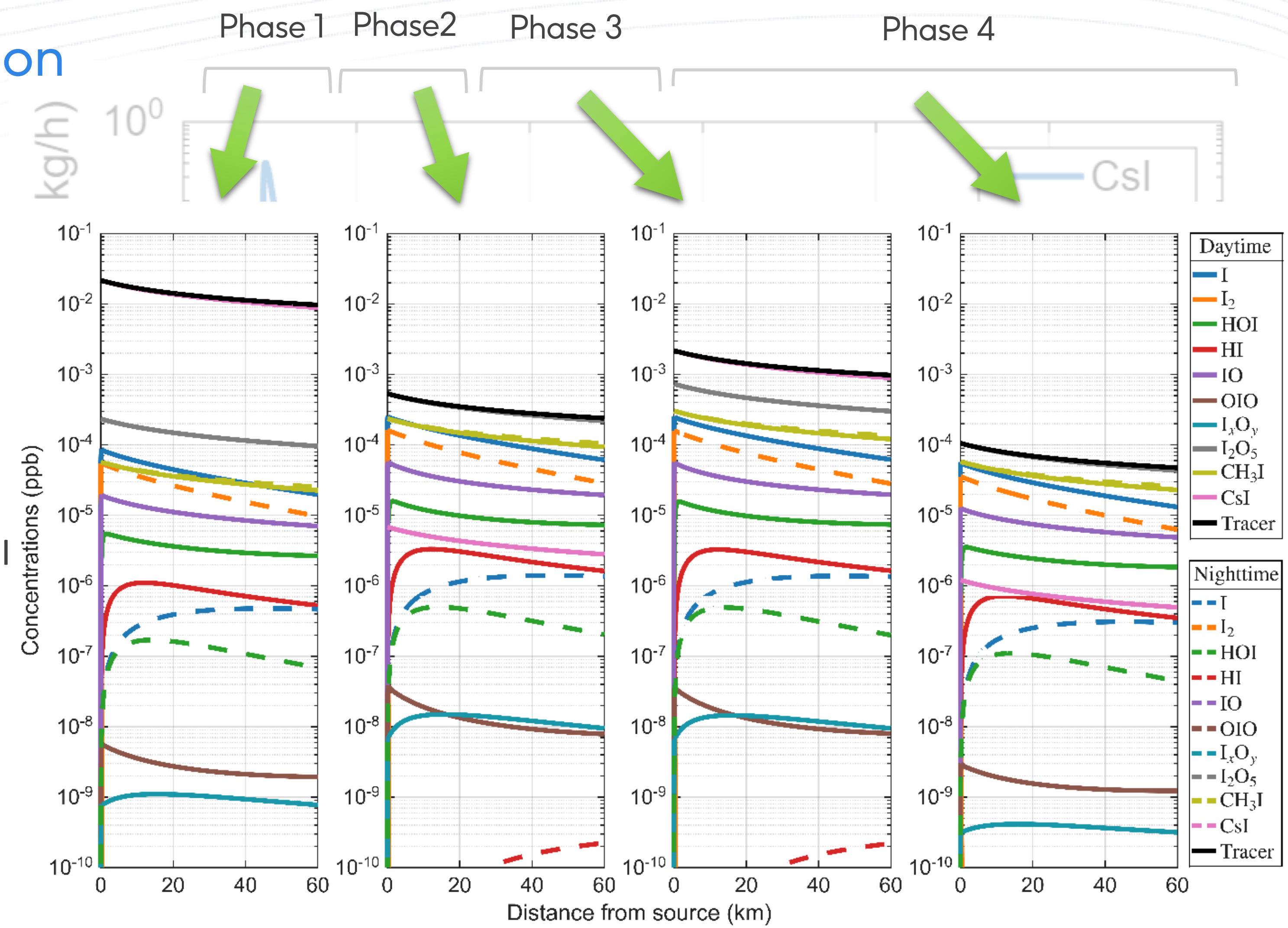

Figure: Atmospheric radioiodine concentrations (Case 1).

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Amalgamated Inhalation Dose Coefficients

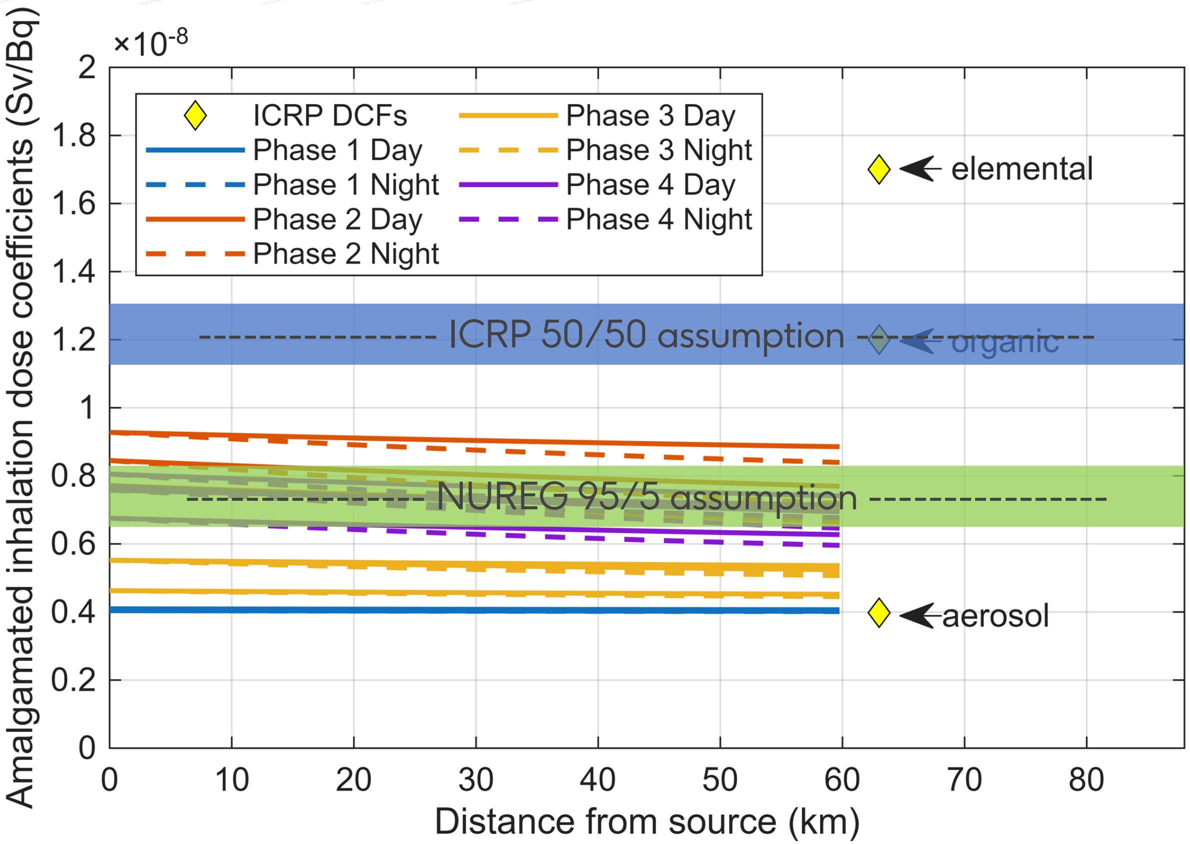

Figure: Amalgamated inhalation dose coefficients.

- Amalgamated DCF based on stoichiometric fraction of species:

$$e_{inh,eff} = \sum e_{inh,i} \cdot f_i$$

- **Phases 1 & 3:** Aerosol-rich and amalgamated DCF ≈ aerosol DCF for 0.2 µm particles.
- **Phases 2 & 4:** Higher DCFs and stronger day/night differences due to higher fraction of elemental + organic species.

Ground Deposition

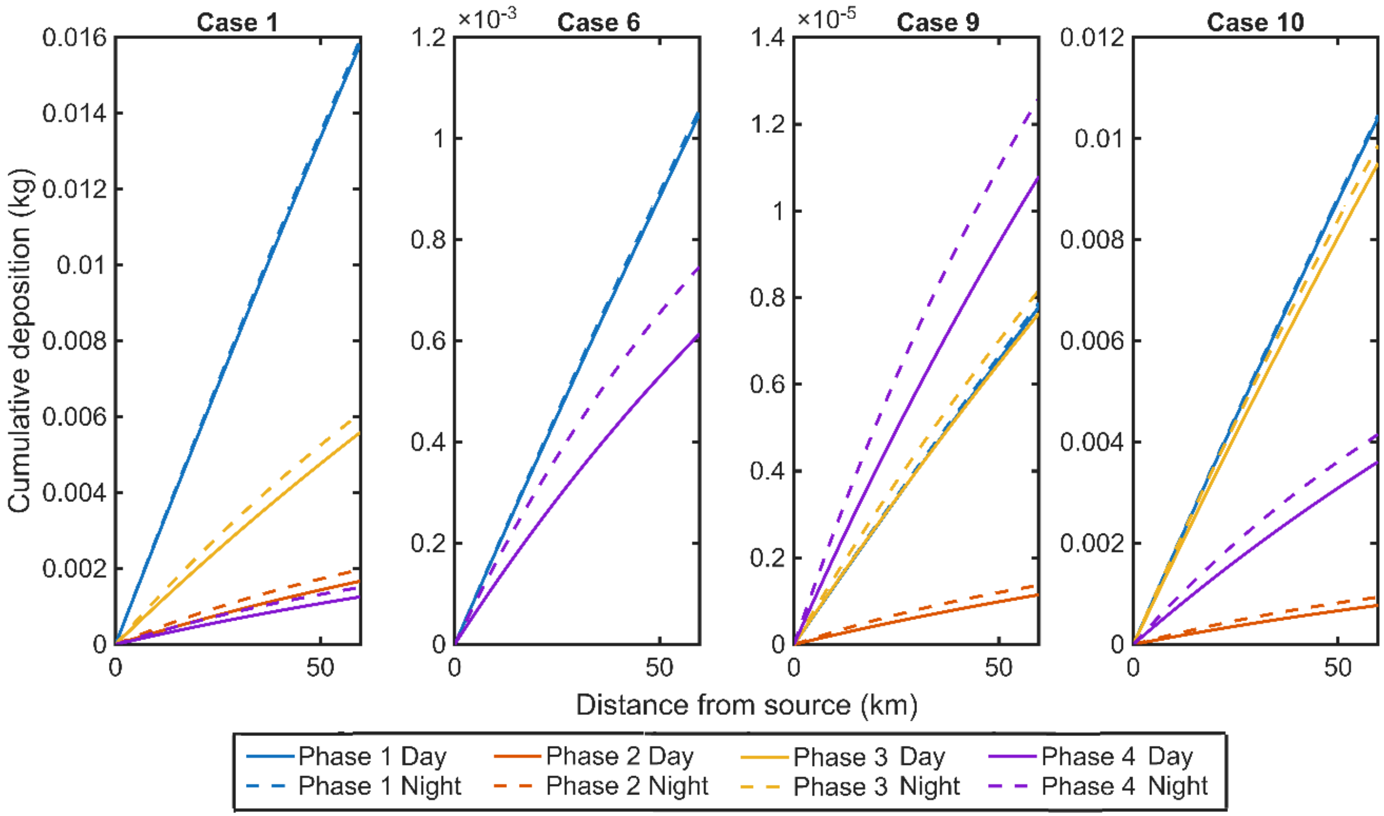

Figure: Cumulative deposition in different cases.

- **CsI aerosol** typically most important for deposition in cases with large releases.
- **Radiolysis products** can be more important in some cases due to higher deposition velocity, longer duration of releases.

Conclusions

- High-fidelity chemistry modeling framework developed integrating ***source term + atmospheric transport***.
- Early accident releases are dominated by **CsI aerosols**.
- Persistent releases (into long term) driven by **radiolytically generated I_2 vapor, organic iodine vapor, and I_2O_5 aerosols**.
- Atmospheric photochemistry and deposition significantly modify the gas-phase iodine fraction, producing strong day-night differences.
- Mixture-weighted inhalation dose coefficients dominated by particulates -> implying ICRP assumptions overly conservative.

Thank you

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