Long-Term Monitoring of Radionuclides in Soils and Groundwater: Lessons Learned from Chernobyl

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Outline

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Motivation

“We don’t need to guess--we know what happened at Chernobyl.”

Skip Chamberlain, DOE
Motivation

- Evaluation of hydrogeological and geochemical information to describe how natural and engineered barriers may degrade
  - Retardation of radionuclides in the unsaturated and saturated zones
  - Degradation of engineered barriers

- Sensitivity studies and validation of the Total System Performance Assessment models

- Decommissioning of Nuclear Power Plants
  - Control of hot particle contamination at nuclear power plants during fuel reconstitution
Chernobyl Exclusion Zone is an area surrounding the site that was sealed off as an institutional control to prevent further access after the accident.
Background Setting

**Chernobyl Region:**
Climate is humid with mild, short winters having frequent thaws, and warm summers

- Average annual precipitation from 550 to 750 mm/year
- Average annual temperature from 5°C to 7°C
  - min monthly average -35°C
  - max monthly average 39°C
- Groundwater depth is 2-3 m

**Southern Ukraine:**
Climate is semi-arid

- Groundwater depth is 50–70 m
I. Water levels
1. Cenomanian Albian aquifer: (a) prior to water intake, and (b) under present conditions;
2. Jurassic aquifer: (a) prior to water intake and (b) under present conditions.

II. Aquifers.
3. Oligocene Quaternary aquifer (sands, sandy loams);
4. Eocene aquifer (sands);
5. Cenomanian Albian aquifer (limestones, chalks, sands, sandstones, gravel);
6. Jurassic aquifer (limestones);

III. Aquitards
7. Quaternary loams and clays;
8. Kiev (Eocene) marls;
9. Turonian marls;
10. Jurassic clays and clayey limestones;
11. Boreholes of the Kiev water intake

Are aquitards impermeable?
Causes of the Chernobyl Accident -- April 26, 1986

1. Loss of water coolant caused the water-circulation system to fail. Temperature in the reactor core increased to over 5,000°F. Uranium fuel melted and produced steam that reacted with the zirconium alloy cladding of the fuel rod to produce hydrogen gas.

2. The reaction between steam and graphite produced free hydrogen and carbon oxides. This gas combined with oxygen, a blast blew off the top of the building, igniting the graphite.

3. The burning graphite threw radioactive fission products into the air.

From *Time Magazine*, 1996
Radionuclide inventory:

- ~5% of the radionuclides from Chernobyl’s 4th unit were released to the atmosphere, with a total activity of $\sim 11 \times 10^{18}$ Bq ($3 \times 10^8$ Ci).

- $\sim 6 \times 10^{17}$ Bq ($1.6 \times 10^7$ Ci) of long-lived radionuclides remained in the destroyed reactor.

- The kinetic energy released was equivalent to 30–40 tons of trinitrobenzene (TNT).

- Contamination was equivalent to that of a 12-megaton nuclear explosion.
Relatively short-lived isotopes of iodine, cesium, and strontium were released.

Long-lived transuranic elements such as plutonium and americium were released

- \(~2,000\, \text{Ci of }^{239,^{240}}\text{Pu}\)
- \(1,344\, \text{Ci of Pu activity was outside the Exclusion Zone}\)

Bondarenko 1998; Bondarenko et al. 2000
Types of Released Radioactive Particles

- **Fuel components** of finely dispersed fuel particles
  - Elements of low volatility, such as cerium, zirconium, barium, lanthanum, strontium, and the actinides
  - Settled primarily within the ChEZ

- **Condensed components**
  - Formed when radioactive gases (iodine, tellurium, cesium, and strontium, and ruthenium) ejected during the nuclear fuel fire
  - Settled primarily along the atmospheric flow pathways, including outside of the ChEZ
Hot Particles

- Formed from both condensed and fuel particles fused with the metal cladding of the fuel rods, and mixed with sand or concrete
- Size from 1 to 100 µm
- Activity concentration $>10^5$ Bq/g
- The surface density of hot particles was $\sim 1,600$ per m$^2$ in downtown Kiev
- Detected in soils to depths of about 0.5 m in 1987 (Gudzenko et al. 1990)
- Included particles of uranium dioxide
In the near-ChNNP zone in 1986:

- <25% of activity was associated with exchangeable forms of $^{90}\text{Sr}$ and $^{137}\text{Cs}$
- The remaining activity was caused by nonexchangeable forms of these radionuclides associated with fuel particles (Kashparov 2001).

- $\beta$-activity of hot particles is caused by $^{90}\text{Sr}$, $^{106}\text{Ru}$, $^{134}\text{Cs}$, $^{137}\text{Cs}$, $^{144}\text{Ce}$, and $^{147}\text{Pm}$.

- $\alpha$-activity of hot particles is caused by actinides ($^{238}\text{Pu}$, $^{239}\text{Pu}$, $^{240}\text{Pu}$, and $^{242}\text{Cm}$).
Difference between Chernobyl and Nuclear Bomb Particles

Chernobyl hot particles:

- Did not contain activation products such as $^{60}\text{Co}$
- Contained $^{125}\text{Sb}$ and $^{144}\text{Ce}$, which are absent in atomic bomb hot particles
- Contained a larger fraction of $^{137}\text{Cs}$ than that in nuclear bombs
- Had a ratio of $^{154}\text{Eu}/^{155}\text{Eu}$ about 10 times greater than that in atomic bomb particles and about 200 times than that in fusion bomb particles
- Have a lower radiation of uranium and neptinium than that from fusion bombs
- Had a lower plutonium content than that in fusion bombs
- Had a much lower activity of $^{90}\text{Sr}$ and $^{137}\text{Cs}$ in water-soluble and exchangeable forms than that in global fallout after nuclear weapons testing

Zheltonozhsky et al. 2001
Atmospheric Pathway Fallout Trajectories in April-May 1986
Radioactive Contamination in Europe
Concentration of $^{90}$Sr and $^{137}$Cs in Water from the Dnieper River Reservoirs

Bottom sediments sorbed most of the radionuclides!

1: Mouth of Pripyat; 2: Kiev Reservoir; 3: Kanev Reservoir; 4: Kremenchug Reservoir; 5: Dneprodzerzhinsk Reservoir; 6: Zaporozh'e Reservoir; 7: Kakhovka Reservoir
Surface Water Contamination

90Sr Concentration vs. Discharge at the Mouth of the Borschi Stream

Reconstruction of the 90Sr flux in the Borschi stream

A Satellite Image of the Chernobyl NPP area

The ChNPP cooling pond:
- Heavily contaminated
- Represented one of the major sources of 90Sr migration

Freed et al., 2004
• $^{90}\text{Sr}$ was ejected with fuel particles and precipitated mainly in the near ChNNP zone.

• The $^{90}\text{Sr}$ content is $\sim 810 \text{ TBq} \ (8.1 \times 10^{14} \text{ Bq})$ within Ukraine, which is 3 to 4 times lower than previous estimates.

• $^{137}\text{Cs}$ was contained in the condensed component, which moved and precipitated further away from the ChNPP.
Map of $^{239+240}\text{Pu}$ (kBq/m$^2$) Density at the Land Surface within the ChEZ in 2000
Vertical velocity of 137Cs migration was 0.4 to 0.7 cm/yr in the first years after the accident, and then dropped to 0.2 cm/yr.

• ~99% of plutonium accumulated within the top 2 cm layer.

Vertical Distribution of $^{90}$Sr in Soils at ~2 km from CNPP along the Northern Trace, 1996.
In Chernobyl soil samples, the \( ^{238}\text{Pu}/^{239,240}\text{Pu} \) ratio varied from 0.25 to 0.35, which corresponds to the ratio of about 0.3 in spent fuel.

For comparison, the ratio \( ^{238}\text{Pu}/^{239,240}\text{Pu} \) for global fallout from atomic bomb testing was from 0.03 to 0.05.

Bondarenko et al. 2000
Dynamics of the $^{137}$Cs Concentration Introduced Into Soil in a Water-Soluble Form

Types of Soils:
1 - Soddy-podzolic soil (arable land);
2 - Soddy-podzolic sandy-loam soil (arable land after liming in very high doses);
3 - Soddy-podzolic gleic sandy-loam soil (arable land);
4 - Soddypodzolic sandy-loam humous soil (meadow);
5 - Soddy-podzolic sandyloam soil (arable land)

Ivanov and Kashparov, 2004
Concentration of $^{90}\text{Sr}$ and $^{137}\text{Cs}$ for Condensed (Dashed Lines) and Fuel (Solid Lines) Components, Depending on pH

Kashparov, 2004
During the first 5-6 years after the accident, the $^{137}$Cs concentration decreased rapidly.

The effective environmental half-life period of $^{137}$Cs:

- 0.7–1.5 years in vegetables and animal agricultural products
- 1.6–4.8 years in milk and other products
- 2–7 years in grains and potatoes (Bruk et al., 1998)
- 1–4 years in fish, water, and terrestrial vegetation during the first five years after Chernobyl, and then increased to 6–30 years since 1990 (Smith et al., 2000).

Use physical values of radionuclide half-lifes gives conservative estimates of radionuclide migration in the subsurface.
Atmospheric resuspension of radionuclides is a secondary source of contamination.

The main cause for the resuspension of radionuclides is wind erosion of soil.

Especially hazardous in agricultural areas.

The resuspension factor was higher immediately after the accident.

Important in determining inhalation (i.e., lung) dose.
Sources of radionuclide contamination of groundwater

- 800 radioactive waste disposal sites within the ChEZ
- The Chernobyl cooling pond
- Migration through and around casings of boreholes
- Migration in colloidal and dissolved states from the vadose zone

$^{90}\text{Sr}$ in a Quaternary aquifer (depth of 7--20 m) in 1996; Kiev industrial municipal agglomeration

$^{90}\text{Sr}$ in an Eocene aquifer (depth of 30--60 m) in 1996
Groundwater Contamination

- $^{134}$Cs in groundwater and the increase of $^{90}$Sr concentration (from 4 to 400 mBq/L) indicate groundwater contamination at 50 to 70 m depths over five years (1987--1992) (Bar'yakhtar 1997).

- At 22 months after the accident, $^{137}$Cs and $^{90}$Sr were detected in the Cretaceous aquifer at depths of 80 to 120 m within the 30 km zone.

- Elevated concentrations of $^{90}$Sr and $^{137}$Cs were found in all aquifers down to 200–300 m.
Water from the Dnieper River is used for irrigation of more than 1.8 million hectares in Ukraine.

Irrigation in the southern Ukraine leads to the accumulation of radionuclides in the topsoil layer and in crops.

The data qualitatively confirm the concept of a radionuclide buildup in irrigated soils that is employed in the Yucca Mountain Biosphere Process Model.
Main mechanisms controlling radionuclide transfer to plants:

- Direct deposition on plant surfaces
  - from atmospheric resuspension of contaminated soil
  - from irrigation with contaminated water
- Root uptake of radionuclides.

The $^{90}\text{Sr}$ transfer coefficient increased from $1 \times 10^{-3}$ to $1 \times 10^{-2}$ m$^2$/kg from 1988 to 1994, because $^{90}\text{Sr}$, which settled on soils, is gradually transformed into a soluble form and thus absorbed by plants.
Interaction between biota and *micromycetes* fungi destroyed hot particles after the Chernobyl accident.

Two mechanisms are responsible for these processes:

- Microbial metabolic activity
- Mechanical destruction by mycelial overgrowth of radioactive particles
Sarcophagus Was Erected in 1986 as an Interim Measure

The activity of stored radioactive waste is $2.4 \cdot 10^{20} \text{ Bq}$
The SIP aims are:

- Stabilization of the existing Sarcophagus
- Construction of a new Shelter around the Sarcophagus
- Confinement of the radioactive substances inside for at least 100 years
- Removal of remaining radionuclides from inside if needed
- Dismantling of the old structure
The IRL

- Established on July 22, 1998, according to the agreement between the governments of the USA and Ukraine.
- Provides an operational facility for the USA personnel, conducting research within the ChEZ:
  - Texas Tech University
  - Savannah River Ecology Lab

DOE provides funds for the equipment, supplies, and limited personnel.

DOE delegation led by Gerald Boyd and Skip Chamberlain visited IRL in September 2001
Examples of Potential Collaboration Areas Using the IRL

- Natural attenuation and bioremediation of radioactive contaminants in soils and groundwater
- Radionuclide migration in soils and groundwater due to irrigation in Southern Ukraine
- Deactivation of transuranic elements
- Research on colloidal transport, absorbents, and bioaugmentation
- Reutilizing of liquid wastes
- Heat and mass transfer under the "Shelter"
- Nuclear Data Base
- Nondestructive methods for the determination of Pu, Am, Sr in water, soil samples, and hot particles, using alpha-, beta-, and gamma spectrometers and fluid scintillation spectrometer
- Comparison of contamination resulting from Chernobyl with that at DOE sites
- Data and models of migration of radionuclides in the chain "fallout-soils-plants-animals-humans," as well as in soils and groundwater
Vadose Zone Investigations

Map of the Chernobyl Pilot Site developed according to the Ukraine-France project

After D. Bugai et al., 2003

Comparison of surface radar tomography and Radon/Thoron isotopic methods to determine zones of preferential flow

Shestopalov et al., 2000
Conclusions

Numerous data collected over the past 18 years after the Chernobyl accident in different parts of the biosphere can be used to:

• Build confidence in DOE and NRC conceptual and numerical models for long-term monitoring and predictions of transport processes and radiation doses
• Assess the risk associated with the use of contaminated groundwater as the source of drinking water, irrigation, animal watering, and domestic uses
• Assess the performance of nuclear power plants within a regulatory framework