Residual radioactive contamination at the Chernobyl site, and prospective of an effective monitoring and remediation assessment program

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Rockville, MD
March 10, 2015

Outline

- **Introduction:**
  - **Challenges to responding to nuclear accidents**
  - **Chernobyl-background information**
- **Atmospheric pathwa EM121 d forms of radioactive releases**
- **Terrestrial, soil, and groundwater pathways of radionuclides**
  - **Depth distribution in the vadose zone and phenomena of preferential flow**
  - **Groundwater vulnerability—fast flow pathways and secondary contamination**
- **Aquatic pathways of radionuclide transport**
  - **Small rivers and watersheds, the Dnieper River**
  - **Chernobyl Cooling Pond (monitoring and decommissioning program)**
- **Ecological and effective half-life of radionuclides and K_d**
- **Examples of monitoring programs**
- **Transferring knowledge gained from Chernobyl, recommendations, and concluding notes**

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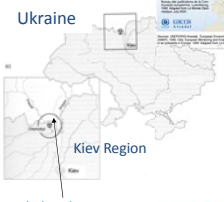
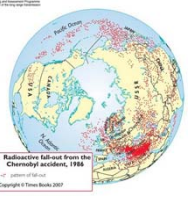
Chernobyl—Background Information

Ukraine





Chernobyl Exclusion Zone was initially assigned within the 30-km radius zone, and then adjusted

http://thewatchers.adorraeli.com/2011/03/16/are-we-facing-a-new-chernobyl-disaster/

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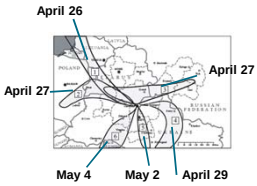
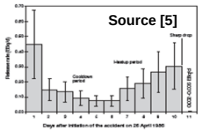
Slide 2

EM121 Depth distribution...
DOE-EM, 1/29/2015

1 Boris Faybishenko, 2/5/2015

Atmospheric pathways of initial radioactive releases





Physico-chemical forms of deposited radionuclides

- **Fuel particles**—finely dispersed, low volatility, settled primarily within the ChEZ
- **Condensed components**—from radioactive gases, settled primarily along the atmospheric flow pathways
- **Hot particles**—fuel particles, uranium dioxide, specific activity >10^5 Bq/g, 1 to 100 µm, surface density ~ 1,600 per m², ~ 0.5 m depth
- **Deposition**--aerosols fall onto the ground due to the gravity or with rainfall. The major part of radioactivity in Western Europe was associated with particles of <2 µm.
- **Resuspension**--aerosols on the ground are lifted in the air by the wind or some other reasons.

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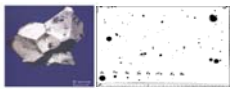
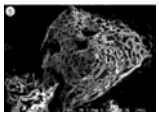
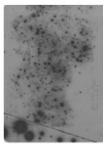
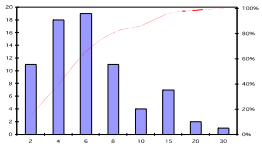
Hot particles resealed from the Chernobyl NPP:

fuel particles, uranium dioxide, specific activity >10^5 Bq/g, 1 to 100 µm, surface density ~ 1,600 per m², detected to ~ 0.5 m depth









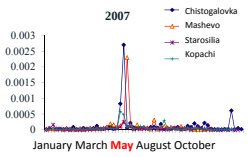
Autoradiography of CP sediments, Ukrainian Institute of Agricultural Radioecology

5

Post-accident sources of contamination in the ChEZ

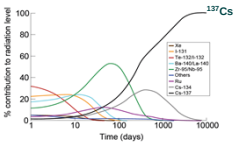
- Planned and unplanned emissions of Chernobyl NPP,
- Excavation of contaminated soil,
- RAW management activities,
- Radionuclides redistribution by wind and fires

Annual increase in ^{137}Cs concentration in plant pollen



Source: Chernobyl Ecocenter

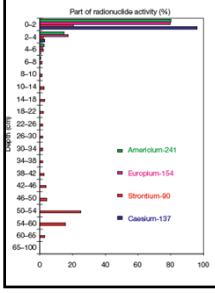
Contributions of the various isotopes to the external (atmospheric) absorbed dose



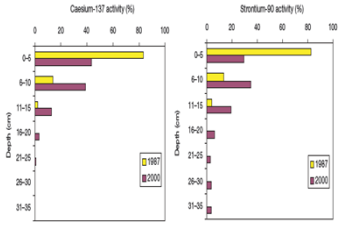
http://en.wikipedia.org/wiki/Chernobyl_disaster#mediaviewer/File:AirDoseChernobylVector.svg

Vertical profile of radionuclides in the vadose zone is affected by preferential flow and transport in topsoil zone

Radionuclides profiles in relatively dry sandy soil in 1996



^{137}Cs and ^{90}Sr profiles in 1987 and 2000 in a soddy gley sandy soil, Gomel region, Belarus



ENVIRONMENTAL CONSEQUENCES OF THE CHERNOBYL ACCIDENT…, IAEA, Vienna, 2006.

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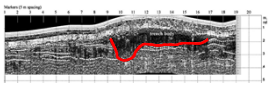
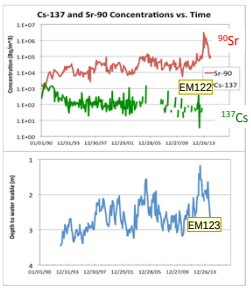
In contrast to the surface water, ^{90}Sr concentration in groundwater continue to grow due to releases from waste disposal sites

800 Radioactive Waste Disposal Sites are significant sources of contamination



The GPR survey to detect the underground trench containing radioactive materials. Red solid line – the trench boundary.



Secondary radionuclide contamination of groundwater. Example from Monitoring Well 2/1 in Red Forest, 1.5 km SW from ChNPP, caused by an increase in the water table.

Challenging problems of Chernobyl Cooling Pond decommissioning and remediation, one of the largest self-contained water reservoirs

Area ~ 22 km^2 , ~1.5 × 10^8 m^3 of water

Water is pumped from the Pripyat River into the Cooling Pond to replenish water lost to seepage and evaporation

Sources of Contamination

- Dispersed fuel particles
- Heavily contaminated water from the reactor basement and soils.
- Total radioactivity >200 TBq, including ^{137}Cs-80%, ^{90}Sr-10%, ^{239,240,241}Pu-10%
- Rel EM125 f contaminated water into the pond

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Slide 8

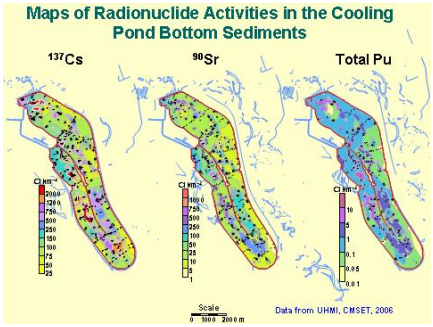
EM122 Difficult to see the white line
DOE-EM, 1/29/2015

EM123 Changed to red for contrast
DOE-EM, 1/29/2015

Slide 9

EM125 Is this still true? Also maybe mention large seepage so needs to continually be filled.
DOE-EM, 1/29/2015

Spatial distribution of radionuclides in bottom sediments is highly variable, depending on the types of sediments and radionuclides



10

Effective radionuclide half-lives in terrestrial ecosystems, years

Process	^{137}Cs	^{90}Sr
Surface run-off	>1000	>300
Descending migration	60–>1000	25–45
Immobilization	0.5–7.0	13–70
Radioactive decay	30	29

Soil	^{90}Sr				^{137}Cs			
	Meadow (0-5 cm)		Arable Land (20 cm)		Meadow (0-5 cm)		Arable Land (20 cm)	
	T_{ecol}	T_{eff}	T_{ecol}	T_{eff}	T_{ecol}	T_{eff}	T_{ecol}	T_{eff}
Automorphous Mineral Soils of Light Granulometric Composition	11–19	6–8	21–32	12–15	60–150	20–25	26–45	14–18
Automorphous Mineral Soils of Heavy Granulometric Composition	15–32	10–15	37–129	16–23	150–400	25–27	60–150	20–25
Hydromorphic Organogenic Soils	100–160	22–24	103–233	22–25	11–20	8–12	30–60	15–20
Drained Organogenic Soils					17–83	11–22		
Hydromorphic Mineral Soils					45	18		
Weakly Humus Sand	2.2–6.0	2–5	2.2–7.6	2–6				

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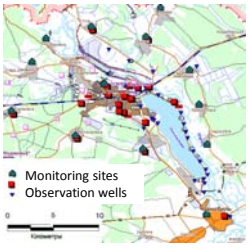
Existing monitoring network is insufficient to obtain detailed information for decommissioning and remediation of the Chernobyl Cooling Pond

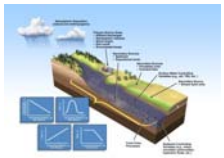


Monitoring needs

- Monitoring of the exposed bottom sediments
- Monitoring of the near surface atmospheric layer
- Groundwater monitoring
- Monitoring of the local ponds developed due to the drop of the water table in the Cooling Pond
- Hydrobiological and geobotanical observations
- Meteorological monitoring

DOE 'Systems-Based' Monitoring Approach

- Integrated systems-based approach to monitoring
  - Contaminated sites a complex systems
  - Compliant, scientifically defensible, cost-effective monitoring approach
  - Guided transition of characterization and remediation phases with the goals of meeting end states
  - Iterative feedback and using multiple "lines of evidence

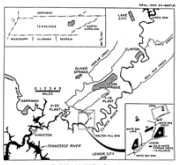


Chernobyl Cooling Pond experience can be used at DOE Sites

Savannah River PAR Pond

- **Constructed in 1958 as a Cooling Pond for the Savannah River Site's P and R Reactors**
- **After the water level drawdown in 1991:**
  - –Exposed 5.3 km² of contaminated sediments.
  - –Mobility of the radionuclides was much greater than expected (Hinton et al. 1999; Whicker et al. 1999).
  - –Cs-137 concentrations increased, probably resulting from the Cs-137 increase in sediments.
  - –Sedimentation rates and water turbidity increased due to erosion processes.

Oak Ridge Reservation--White Oak Lake



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Concluding Notes: Chernobyl as a Case Study for Evaluating Residual Radioactivity in the Subsurface at Operating and Decommissioning Nuclear Power Plants

- Chernobyl is an inadvertent laboratory to take advantage of almost 30 years wide-spread radioactive contamination data:
  - to better understand the challenges in responding to a nuclear reactor accident
  - to study processes affecting transport and natural attenuation of exposed and subsurface radionuclides,
  - to test wide range of monitoring, modeling, risk assessment, data management, and design technique by US DOE, US NRC, and other US Federal and International agencies.

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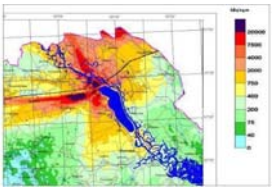
Background slides

Long-term effects and hazards of wind resuspension



Although wind resuspension of radioactive particles can be a secondary source of contamination for areas not exposed to the original release, modeling shows that the radionuclide resuspension by wind is negligible in comparison to the initial post-accident radioactive contamination (Kashparov et al., 2001)

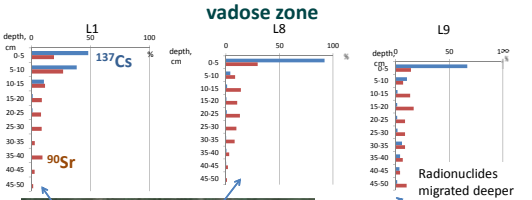
EM126

Hazardous resuspension due to fires in the Chernobyl Exclusion Zone and the surrounding area of obligatory resettlement

- The NOAA Advanced Very High Resolution Radiometer satellite image of a smoke plume originated in the Chernobyl Exclusion zone.
- The plume traveled ~100 km and reached the city of Kiev.

Hao et al., 2009.

Near-surface ^{137}Cs and deeper ^{90}Sr vertical profiles in the vadose zone



Radionuclides migrated deeper near the Chernobyl Cooling Pond

Chernobyl Cooling Pond

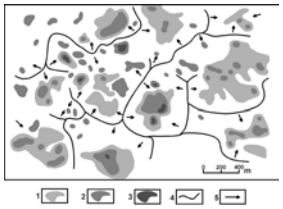
Source: data collected by Chernobyl Ecocenter in 2011

Slide 17

EM126 Delete extra "l"
DOE-EM, 1/29/2015

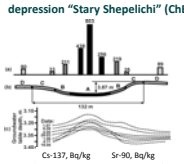
Groundwater vulnerability is affected by the presence of local topographic depressions and phenomena of fast radionuclide transport toward groundwater

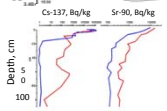
Accumulation of radionuclides in local topographic depressions



Scheme of depression occurrence at a site of in the ChEZ. Depressions with depths to: (1) 0.5 m, (2) 1.0 m, (3) 1.5 м, (4) watershed dividing lines, (5) surface water flow directions.

Increased annual infiltration in the depression "Stary Shepelichi" (ChEZ)



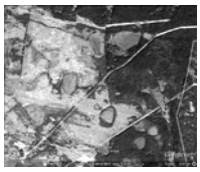


EM127

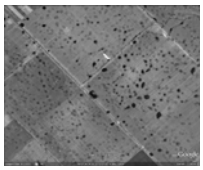
^{137}Cs and ^{90}Sr depth distribution, Veresok site (Shestopalov, 2009)

19

Local surface depressions causing fast flow and transport toward groundwater can be seen from areal photographs



Ukraine, ChEZ, 20 km W from Chernobyl NPP

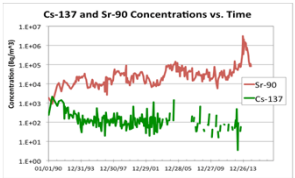


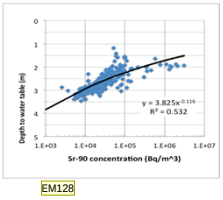
Ukraine, Chernigiv oblast, Yablunivka, 18 km SW from Pryluki (E from ChEZ)

ChEZ depressions may capture up to 60–80% of surface runoff (Shestopalov et al., 2014)

20

Secondary Radionuclide Contamination of Groundwater in Monitoring Well 2/1 in Red Forest, 1.5 km SW from ChNPP, caused by an increase in the water table





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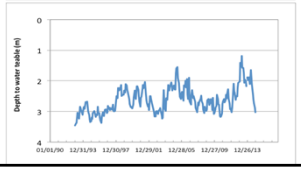


Similar increase in Sr-90 concentration in groundwater was observed in other monitoring wells in 2013-2014

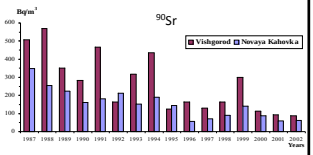
Slide 19

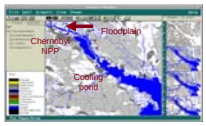
EM127 Figure too small to read, so make sure to define the x and y axis
DOE-EM, 1/29/2015

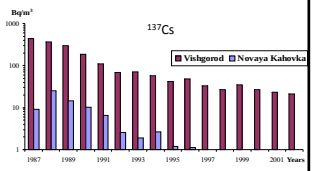
Slide 21

EM128 Identical or Similar trend of increasing…
DOE-EM, 1/29/2015

^{90}Sr and ^{137}Cs in the Dnieper River water reservoirs decreased with time



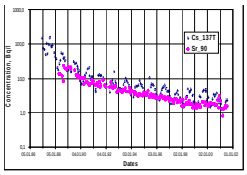




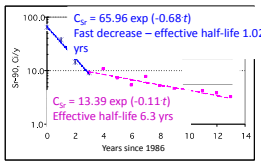
^{90}Sr concentration in the Dnieper River is still above of its pre-accidental levels
^{137}Cs concentration in the lowest reservoir returned to its pre-accidental level in 1996-1998
After Voitsekhovich and Zheleznyak.

Evaluation of the Ecological and Effective Half-Life of ^{137}Cs and ^{90}Sr, based on time trends of concentrations in the Chernobyl Cooling Pond

EM129



Data after O. Nasvit



$$N(t) = N_0 \left(\frac{1}{2}\right)^{t/t_{\frac{1}{2}}}$$

$$N(t) = N_0 e^{-\frac{t}{\tau}}$$

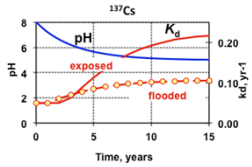
$$N(t) = N_0 e^{-\lambda t}$$

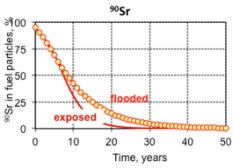
N_0 is the initial concentration that will decay,
$N(t)$ is the remaining concentration at time t,
$t_{1/2}$ is the half-life of the decaying quantity, called mean life-time of the decaying quantity,
λ is a positive number called the decay constant

23

K_d parameters of bottom sediments depend on redox conditions

- Low DO and high pH cause a very slow dissolution of fuel particles in bottom sediments.
- Vegetation and microbiological activity will acidify newly formed soils, causing the dissolution rate to increase.
- Fuel particle dissolution will take ~15–25 years in exposed sediments, and ~100 years in flooded areas (Bulgakov et al., 2009)
- K_d depends on the *N* ammonia concentration (M. Zheleznyak et al., (2005)—INTAS-2001-0556 Project Report "Radionuclide and Sediment Transport Modelling Within the Cooling Pond Ecosystem")





A. Konoplev et al. 2009

Slide 23

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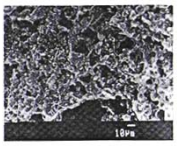
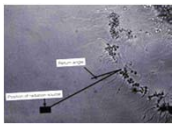
DOE-EM, 1/29/2015

Wide variety of microbiological activities detected: Filamentous Fungi, Survivors of Chernobyl

- **Fungi isolated from the Chernobyl site showed a tendency to grow toward hot particles, overgrow and dissolve them over time.**
- **Accumulation of radionuclides from intact hot particles more intensive for ^{152}Eu than for ^{137}Cs by a factor of about 2.6–134**
- **Cs and Eu accumulation in fungi influenced by**
  - **fungal species and strains**
  - **size and composition of hot particles**





Nelli Zhdanova, et al., Institute of Microbiology and Virology, Kyiv, Ukraine
John Dighton, Rutgers University, USA
Mykola Kuchuk, et al., Institute of Cell Biology and Genetic Engineering, Kyiv, Ukraine
Tamas Torok, Center for Environmental Biotechnology, Lawrence Berkeley National Laboratory, USA

Microbiological Processes: Collection of filamentous fungi

- Some 2,000 filamentous fungi were collected in the ChEZ
- About 25% of fungi showed **positive radiotropism** upon isolation
- Directional growth of fungal mycelium towards the source of low-level ionizing radiation
- Independent of the nature of radiation
- 86% of fungi isolated from radioactively contaminated sites showed positive directional growth while control isolates from pristine sites were negative
- Biological species, exposed to the tremendous radiation in and around the failed nuclear plant, may experience a "speeded up" evolution
- Fungal biomass may be used for bioremediation of radionuclides

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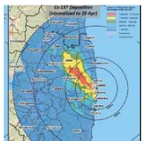
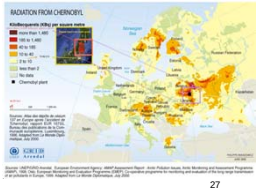
US-DOE Supported Investigations at Chernobyl and Fukushima

- DOE Comprehensive Chernobyl database
- REM - Radioactivity Environmental Monitoring databank (over 2 million data records)
- EURDEP - European Radiological Data Exchange Platform
- ECRIN - Dose coefficients to calculate ionizing radiation doses received by humans.
- "Seeds in Chernobyl" - Plants database
- INIS - International Nuclear Information System Database.
- RESPOND— Database of Radio-Ecological Study
- RODOS – Data Management System used at Chernobyl and Fukushima



Aerogamma Measurements by MEXT and DOE



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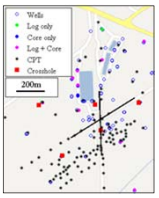
ASCEM is a state-of-the-art DOE approach, using an integration of toolsets for understanding and predicting contaminant fate and transport in natural and engineered systems.



Uranium-Contaminated Savannah River Site, USA

http://ascemdoe.org/

U.S. Department of Energy, BER

Scientific Focus Area Field Research Site at Rifle, Colorado

Site Operations: U.S. Department of Energy, Legacy Management, Grand Junction, Colorado

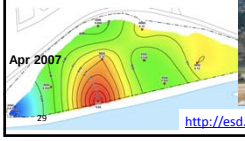
Site Owner: City of Rifle ***Site size:*** 22 Acres

Site History: Started in 1924 (Radium and Vanadium), Uranium milling during the 1950's

Key site issues: What is limiting natural attenuation of U? What controls subsurface redox, C and N cycling and fluxes in flood plains?

~1957 Uranium mill (shut down Dec 1957)

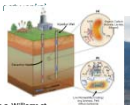
Site today



http://esd.lbl.gov/research/projects/sustainable_systems/

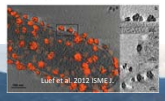
Rifle Site Data Collection and Critical Outcomes

Biogeochemistry (contaminant concentrations/reaction



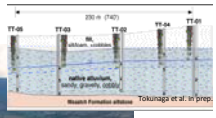
e.g. Williams et al. 2011 Geomicro J.; Long et al. 2015 GCA

Microbiology (redox cycling of metals)



Luef et al. 2012 ISME J.

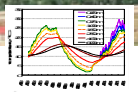
Geohydrology (fluxes)



Tokunaga et al. in prep.

Isotope geochemistry (contaminants sources, mixing models)

Christensen et al. 2014 AGU Fall Mtg



Meteorology, including subsurface temperature (non-isothermal reactions kinetics)

Arora et al. 2014 AGU Fall Mtg

Geophysics (naturally reduced zones in subsurface)

Flores Orozco et al. 2011 JGRB

Examples of monitoring equipment used at DOE Sites

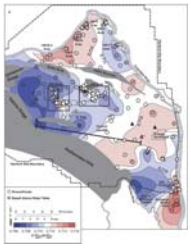
NARAC performs planning, real-time assessment, emergency response, and detailed studies of incidents involving a wide variety of hazards, including radiological releases

NARAC is a distributed system, providing modeling and geographical information tools, real-time access to global meteorological and geographical databases, and advanced three-dimensional model predictions

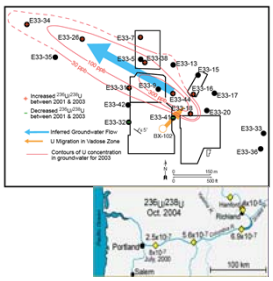
National Center at LLNL

- Advanced, automated 3-D modeling system re-locatable anywhere in the world in real-time
- Scientific and technical staff provides training, assistance, and detailed analysis 24 hours x 7 days.

Hanford isotopic studies to identify plume pathways and rock-water interactions



Sr isotope map of Hanford groundwater



U isotopes to map long-distance plume migration

(Christiansen et al., 2007)

Examples of monitoring equipment used at DOE Sites

Nevada National Security Site

Air Sampler

Thermo Luminescent Dosimeter

Pressurized Ion Chamber

Savannah River Site

2D mapping of soil radioactive contamination using NaI (sodium iodine) detectors

After C.A. Eddy-Dilek and B.B. Looney (SRNL)

Real-time remote radioactivity sensors and monitoring system for soil and groundwater