



Methodology for assessment of exposures to people following nuclear accidents

Gerhard Proehl

Radionuclides released during the Chernobyl accident ENEP (1 PBq = 10¹⁵ Bq)

Volatility	Radionuclide	Half-life	Activity released (PBq)
Noble gases	Kr-85	10.7 a	33
	Xe-133	5.3 C	6500
Volatile elements	Te-129m	33.6	240
	Te-132	<mark>3,3</mark>	<u>1150</u>
	I-131	<mark>8.0 d</mark>	1760
	I-133	20.8 h	<u>91</u> 0
	Cs-134	2.06 a	47
	Cs-136	13.1 d	<u>36</u>
	Cs-137	<mark>30 a</mark>	85
Intermediately volatile elements	Sr-89	50.5 d	115
	Sr-90	29.1	10
	Ru-103	39.3 d	> 168
	Ru-106	368 d	>73
	Ba-140	12.7 d	> 240
Non-volatile elements	Zr-95	64 d	84
and fuel particles	Mo-99	2.75 d	>72
	Ce-141	32.5 d	84
	Ce-144	284 d	59
	Np-239	2.35 d	400
	Pu-238	87.7 a	0.015
	Pu -239	24065 a	0.013
	Pu-241	14.4 a	2.6
	Pu-242	376000 a	0.00004
	Cm-242	18.1 a	0.4





Radionuclide	Release (PBq)		
	Chernobyl (IAEA 2006)	Fukushima (IAEA 2015)	
I-131	1760	100-400	
Cs-134	47	8.3-50	
Cs-137	85	7-20	
Sr-90	10	0.003-0.14	
Xe-133	6500	6000-12000	



Comparison Fukushima – Chernobyl (same scale) ENEP

FALLOUT COMPARISONS

New data from Fukushima show caesium-137 levels approaching those of Chernobyl — but over a much smaller area.



Fukushima compared to Chernobyl: comparable Cs-deposition levels but over smaller area After Vandenhove et al., 2012



Fukushima, 2013/12/17









Radioecological model





ENEP Dose from radionuclides in the passing radioactive plume

Dose = Activity in air × time × dose coefficient × shielding factor (mSv) (Bq/m³) (d) (mSv/d per Bq/m²)





Radiation dose from radionuclides deposited on ground

Dose = Activity on the ground × time × dose coefficient ×shielding factor(mSv)(Bq/m²)(d)(mSv/d per Bq/m²)





ENEP Radiation dose from inhalation during the passage of the plume

Dose = Time integrated activity in air × breathing rate (age) × Dose coefficient(mSv) $(Bq × d/m^3)$ (m^3/d) (mSv/Bq)





ENEP Radiation dose due to intake of food

Dose = activity (food) × food intake × Dose coefficient (mSv) (Bq/kg) (kg/d) (mSv/Bq)



Activity concentrations in food

- Pronounced time-dependence
- Can be determined by monitoring
- Can be predicted by means of radioecological models







Dose coefficients

Convert activity [Bq] in the environment into dose [Sv]

ENEP Dose coefficients

External exposure from radionuclides in the passing plume

- Convert activity per unit air volume (Bq/m³) to a dose rate (Sv/s per Bq/m³)
- Semi-infinite cloud
- No shielding

External exposure from radionuclides deposited on the ground

- Convert activity per unit area (Bq/m²) to a dose rate (Sv/s per Bq/m²)
- Infinite area
- No shielding

Inhalation

- Convert activity per unit activity inhaled (Bq) to a dose (Sv/Bq)

Ingestion

- Convert activity per unit activity ingested (Bq) to a dose (Sv/Bq)

Calculated by ICRP

- Published in a number of ICRP publications 56 (1990), 72 (1996) and 119 (2012)
- Age-groups: 3 months, 1, 5, 10, 15 years, adults
- Values are given for effective dose:
 Weighted mean of doses over all tissues and organs



Biokinetic model for intake of radionuclides with food and drinking water (simplified according to ICRP)



Activity in foodstuffs as function of time after an accident



Which factors are determining activity in foods ?

Radiological characterization	Radionuclides deposited Deposition per unit area Dry deposition or deposition with rain
Environmental characterization	Ability of soils to sorb/fix caesium Agricultural practice (e.g. use of fertilizer) <i>Season of the deposition</i>



Relevant food products

Plant foodstuffs

-Cereals, tubers, vegetables, fruit

• Feedstuffs

- Pasture grass, cereals

Animal foodstuffs

-Milk, beef, pork, chicken, eggs

Food processing and culinary preparation

- -Milk => butter, milk => cheese
- -Cereals => flour
- -Cooking
- -Rinsing



Model processes and parameters

- Key processes following direct deposition on the leaves (1st year)
 - Dry deposition of radionuclides to soil and vegetation
 - Interception of wet deposited radionuclides by vegetation
 - Weathering loss from vegetation
 - Transport of radionuclides in plants to the edible parts

Key processes following deposition on soil (following years)

- Uptake of radionuclides by plants from soil
- Migration and fixation of radionuclides in soil
- Intake of radionuclides by domestic animals
- Transfer of radionuclides to meat, milk and eggs
- Modification of activity in foods during processing and culinary preparation.

ENEP Contamination routes for plant products

A Short-term

- 1 Direct deposition onto edible parts of plants
- 2 Deposition onto leaves -> transport to the edible parts

B Long-term

- 3 Deposition on soil and uptake through the roots
- 4 Resuspension of dust and redeposition on leaves and fruits





ENEP Direct deposition on the plant

Dry deposition

 Radionuclides are removed from the atmosphere without involving precipitation

Dry deposition depends on

- Plant's development (leaf area index)
- Chemical-physical form of the radionuclide
 - Particles size
 - Reactive gases
 - Elemental iodine
 - Sulphur dioxide
- Meteorological conditions
 - -Wet or dry surfaces

Interception

 Radionuclides in rainwater that do not reach the soil, but instead intercepted by the leaves, branches of plants

Interception depends on

- Plant's development (leaf area index)
- Radionuclide and its chemical form
- Amount of precipitation



Dry deposition

Deposition = Time-integrated activity in air * deposition velocity

Deposition velocity $v_g [m/s] =$

Activity deposited on the ground [Bq/m²]/ Time-integrated activity in air $[Bq s/m^3]$

 $-v_{g}$ is determined in experiments

Deposition velocities

- Particles
 - 1 µm
 - 10 µm

- Reactive gases (I₂, SO₂): \approx 0.01 m/s

-Inert gases (CH3-I) \approx 0.0001 m/s

 \approx 0.001 m/s

 \approx 0.01 m/s





Interception of wet deposited radionuclides by plants



- An important process for contamination of plants
 - Rainfall effectively removes radionuclides from the atmosphere
 - -=> Rainfall during the passage of a radioactive plume will increase the total deposition to the ground
- Radioactivity deposited on the leaves are effectively taken up
- Radioactivity deposited on soil
 - Readily sorbed to soil
 - => Low uptake by plants
 - -=> Only relevant for long-lived radionuclides (T1/2 > 1 year)
 - => Loss of radionuclides to deeper soil layers



Dependence of interception fraction on amount of rainfall (Kinnersley et al., 1997)





Dependence of interception fraction on leaf area index (Hoffman et al. 1995)





Interception depends on the chemical form (Hoffman et al., 1995)



(mass interception fraction = interception fraction normalized to the biomass)



Seasonality of growth: Development of wheat

Continuous change of plant morphology during the growing period



Development of leaf area and biomass



MÜLLER, H. PRÖHL, G., ECOSYS 87: A dynamic model for assessing radiological consequences of nuclear accidents, Health Phys. 64 (1993) 232-252.





Stylized development of leaf area index of winter wheat

- Slow development at the beginning of growth
- Rapid growth in spring when temperatures increase



Rainfall and interception

Fraction of activity retained by crops (interception) ...

- -... decreases with amount of rainfall
- -... increases with the development of crops
- -... highest during the peak season



Increase of deposition with increasing rainfall (intensity)







Translocation

Active transport of elements in plants

 Defines the amount of activity transported from leaves to edible parts



Depends on

- Element
 - Mobile elements (xylem + phloem)
 - Immobile elements (only phloem)
- Stage of development
- Pronounced seasonality
- Foliar uptake may exceed root uptake by orders of magnitude



Mobile elements

- Systemic transport in plants from the leaves to other plants organs and vice versa
 - Transport in the phloem
 - Maximum of contamination of the edible part is some weeks before harvest
 - Then, the physiological activity is high, which is the driving force for the transport
- Examples for mobile elements
 - <mark>Cs</mark>, K, Na, S, P, Cl, <mark>I</mark>, Mn, Zn



Immobile elements

- No systemic transport from the leaves to the roots
 - -Only transport in the xylem
 - Only external contamination of edible parts
 - no translocation to potato tubers
 - no contamination of grain before the emergence of ears
- Examples for immobile elements
 - Sr, Ba, Ra, Ce, Pb, lanthanides, actinides



ENEP Translocation factors for wheat and barley



Left: Total activity in grain [Bq/m²] per Total activity deposited on the plant [Bq/m²] Right: Activity concentration in grain [Bq/kg] per Total activity deposited on the plant [Bq/m²]





Activity in edible part of the plant =

(Dry deposition + Wet Deposition * Interception factor) * Translocation factor Yield



Example to calculate foliar uptake of Cs-137 by wheat ENEP following wet and dry deposition

Dry deposition: 1000 Bg/m² Cs-137

 Translocation: Fraction of caesium which is transported from the leaves to the grain (@ 4-8 weeks before harvest): ~ 0.1

• Activity_(wheat) =
$$\frac{1000 \frac{Bq}{m^2} * 0.1}{0.5 \ kg/m^2}$$
 = 200 Bq/kg (dry deposition)

 $0.5 ka/m^2$

Wet deposition: 1000 Bq/m² Cs-137

- Interception: Fraction of activity deposited which is retained by the plant ~ 0.1 for a rainfall of 10 mm (equivalent to 10%)
- Translocation: 0.1
- Yield: 0.5 kg/m²

• Activity_(wheat) =
$$\frac{1000 \frac{Bq}{m^2} * 0.1 * 0.1}{0.5 \ kg/m^2}$$

= 20 Bq/kg (wet deposition @10 mm rain

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Loss of radionuclides from plants due to weathering



• Post-deposition activity loss from plants

- -Rainfall, fog, foliar abrasion
- Including the decrease of activity concentration due to increase in biomass (growth dilution)

• Influencing factors

- Time after deposition
 - Loss rate declines with time after deposition
- Age of plants
 - Higher for young plants
- Rainfall, fog
- Quantified by a weathering half-life
 - 14 days represents a reasonable value





Cs-137 and I-131 activity in weed (leafy vegetables) at 36 km NW from FDNPP

• γ -dose rate drops from 26 \rightarrow 6.5 μ Sv/h from 20 March \rightarrow 1 June

- The decline of I-131 is faster due to the shorter half-life
- The Cs-137 in the end of the of the observation period is due to uptake of Cs-137 from soil


Weathering half-lives (days) of selected elements and for generic plant types

Element	Plant group	N	Arithmetic mean	Minimum	Maximum
Cs	Cereal	1	35		
Cs	Grass	4	10	7.9	11.1
Ι	Grass	9	13	8.3	29
Ι	Rice	1	14		
Sr	Grass	4	24	12.8	49
Sr	Cereals	1	21		
Mn-Ce	Cereals	1	30		TECHNICAL REPORTS SERIES NO. 470
Pu	Cereals	1	12		Handbook of Paramete Values for the Predictio
Pu	Fruits	1	43		of Radionuclide Transfer i Terrestrial and Freshwate Environment

^a Including growth dilution.

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Radionuclide uptake from soil



Long-term source of plant contamination, depending on

Soil characteristics

- Sorption capacity (Sand, loam and clay content, Organic matter)
- -pH value
- Redox potential (esp. iodine, plutonium)
- Concentration of antagonists
 - Cs vs K, Sr vs Ca
 - Use of fertilizer
 - Depth of the roots
- Chemical form of the deposit
 - -Soluble vs inert particles
- Time since the contamination
 - Progressing sorption, fixation and incorporation processes



Quantification of the uptake of radionuclides from soil



- Transfer factor soil-plant TF
 TF= Activity in plant/Activity in soil
 [Bq/kg fresh per Bq/kg dry]
- Aggregated transfer factor T_{ag}
 T_{ag} =Activity in plant/Deposition to soil
 [Bq/kg fresh per Bq/m²] = [m²/kg]
- T_{ag} is used when it is difficult to determine the average activity concentration in soil
 - -In forests
 - On natural land





Typical values for transfer factors soil-plant



- Strontium:
 - -0.1 1
- Caesium
 - -Well managed soils: 0.001-0.1
 - -Organic, acid soils: 0.1-10
- Technetium:
 - -0.1 10
- lodine:
 - 0.001 1
- Plutonium, americium :
 - -0.00001 0.001
- Pronounced variability, also on the same site







Transfer factors soil-plant



Advantages

- Easy determination
- Easy application in models

Disadvantage

- Lumped parameter including all processes
 - Chemical forms of the contaminant
 - Soil properties are not considered
- Large data bases are required to ensure reasonable assessment



ENEP Estimation of root uptake of caesium

Root uptake:

- Deposition: 10000 Bq/m²
- Ploughing depth: 0.25 m
- Soil density: 1400 kg/m³

• => $Activity_{(soil)} = \frac{Deposition}{Ploughing depth*soil density} = \frac{10000 Bq/m^2}{0.25 m * 1400 kg/m^3} \approx 30 Bq/kg$

- Transfer factor soil plant (TF_{sp}): 0.01 Bq/kg plant per Bq/kg soil
- => Activity_(plant) = Activity_(soil) * TF_{sp} = **30** Bq/kg * 0.01 Bq/kg plant p. Bq/kg soil \approx **0.3 Bq/kg**





Contamination route	Dry Deposition	Wet deposition (1000 Bq/m ²)		
	(1000 Bq/m²)	5 mm rain	10 mm rain	
Root uptake	0.3	0.3	0,3	
Foliar uptake	200	40	20	

Comparison of foliar and root uptake of Cs-137

- Radionuclides deposited on foliage are usually effectively taken up
- The activity levels in the edible part can be orders of magnitude higher than due to root uptake
- For long-lived radionuclides, root uptake represents a long-term source for the contamination of crops

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Resuspension of soil



- Defines the flux of radionuclides from soil to atmosphere
- Depends on
 - Soil texture and humidity
 - Vegetation cover
 - Wind speed
- Areas particularly affected by resuspension
 - Arid regions
 - In temperate climates, resuspension during storms may cause a relevant activity loss from soil

Importance of resuspension

• Resuspended soil may be redeposited on crops

- Resulting contamination of plants is low
- Contributes to the overall plant contamination only if root uptake is low

• Factors causing higher resuspension

- Important for canopies with low soil coverage
- Primarily important for small soil particles (clay, silt, very fine sand)
- Radionuclides may be enriched in the resuspendable fraction
 - Enrichment of caesium in fine soil fractions by a factor of 3

Quantification as kg soil per kg plant

- -Same dimension as Transfer factor soil-plant
- For temperate climates: 0.01-1 g soil per kg plant



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Transfer to animal products



- Use of contaminated feedstuffs
- Transfer to meat, milk, eggs



Simple model for the transfer to animal products







Animal	Feedstuff	Intake rate (kg/d)
Cow	Grass/hay	≈ 14
Sheep (lactating)	Grass/hay	≈ 1.8
Beet (meat)	Grass/hay, maize	≈ 10
Pig	Cereals	≈ 3
Lamb	Grass/hay	≈ 1
Hen, chicken	Cereals	≈ 0.1

Feed intake of domestic animals

- Feed intake is very variable
- Type of feedstuffs used depends on the availability of feedstuffs
- The dry matter intake depends on milk yield, age, growth of the animal, etc.







Transfer factors feed-animal products

• Definition

TF _{feed-(meat/milk/eggs)} [d/kg]=

= Activity_(meat/milk/eggs) [Bq/kg] / Daily activity_(animal) [Bq/d]

TF is defined for equilibrium conditions

Dependent on

- Element
- Chemical form
- Feeding diet, feeding status

Data base

- Acceptable data base for I, Sr, Cs to milk
- Poor data for most other elements/products
- Dynamic needs improvement
- Impact of stable elements, especially iodine is unclear



Typical values for transfer factors feed-animal products

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Element	Milk (d/L)	Beef (d/kg)	Pork (d/kg)
Caesium	0.003	0.04	0.4
Strontium	0.001	0.0003	0.002
lodine	0.003	0.001	0.003
Plutonium	5E-5	5E-5	3E-4



ENEP Biological half-lives for animal products

Element	Product	Fraction 1 st component	Half-life 1 st component	Fraction 2 nd component	Half-life 2 nd component
Caesium	Milk	0.8	1.5	0.3	15
	Beef	1	50	-	-
	Pork	1	35	-	-
	Lamb, chicken	1	20	-	-
	Eggs	1	3	-	-
Iodine	Milk, eggs	1	0.7	-	-
	Meat, chicken	1	100	-	-
Plutonium	Milk, meat	1	Years	-	-
	Chicken, eggs	1	25	-	-
Strontium	Milk	0.9	3	0.1	100
	Meat	0.2	10	0.8	10
	Chicken	0.5	3	0.5	100
	Eggs	0.5	2	0.5	20

Model Description of the Terrestrial Food Chain and Dose Module FDMT in RODOS PV6.0 RODOS(RA3)-TN(03)06 https://resy5.iket.kit.edu/RODOS/Documents/Public/HandbookV6/Volume3/FDM_Terra.pdf



Model Description of the Terrestrial Food Chain and Dose Module FDMT in RODOS PV6.0

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Seasonality of Cs-137 in milk following the ^{ENEP} Chernobyl fallout in spring 1986 (UNSCEAR 2008, dairy farm near Munich, Germany)



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Food processing and culinary preparation

• Crops can be directly ingested

- Leafy vegetables:
- Fruit vegetables (tomato, cucumber, etc.)
- Fruit (peach, apples, pear, plum, etc.)

• Products may be processed

- Cereals -> bread
- Milk -> butter, cheese
- Quantification
 - Procession factor F_r
 - F_r = Activity_(processed product)/Activity_(raw product)



Processing factors for cereals, vegetables and milk

Raw product	Processed product	Processing factor Activity processed prod/Activity raw product				
		Strontium	Caesium	Iodine		
Wheat	Flour Bran	0.5 3.0	0.5 3.0	0.5 3.0		
Rye	Flour Bran	0.5 3.5	0.6 2.7	0.5 3.0		
Spring barley	Beer	0.04	0.1	0.1		
Potatoes	Peeled potatoes	0.8	0.8	0.8		
Vegetables	Washing. removal of outer leaves	0.8	0.8	0.8		
Milk	Butter Cream Cheese Cheese	0.2 0.4 6.0 0.8	0.2 0.7 0.6 0.6	0.5 0.7 0.6 1.4		

Model Description of the Terrestrial Food Chain and Dose Module FDMT in RODOS PV6.0 RODOS(RA3)-TN(03)06 https://resy5.iket.kit.edu/RODOS/Documents/Public/HandbookV6/Volume3/FDM_Terra.pdf





Raw product	Processing	Processing factor
Brown rice	Milling to white rice	0.46±0.03
	Washing	0.96±0.03
White rice	Washing	0.58
Brown rice	Boiling rice after milling to white rice and washing	0.13±0.01
White rice	Boiling rice after washing	0.27±0.02
	Boiling rice noodle	0.33±0.07

Processing factor = Activity in processed product/Activity in raw product



Tagami et al., 2019



For more information:

Compilation of parameters for environmental transfer TECHNICAL REPORTS SERIES NO. 472

Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments





Intake of radionuclides with foodstuffs

Internal dose from ingestion:

Intake of activity with all foodstuffs considered

$$A_{h}(t) = \sum_{k=1}^{K} C_{k}(t) \cdot V_{k}(t)$$

- $-A_h(t) = human activity intake (Bq/d)$
- -k = number of foodstuffs considered
- $-C_k$ (t) = activity conc. of foodstuff k (Bq/kg)
- $-V_k(t) = consumption rate (kg/d) of foodstuff k$





Which factors are determining the intake of radionuclides with food

Lifestyle

- National, regional and individual habits
- Do people produce their own food ?
 - If yes, to which extent?

Origin of food

- Where does the food come from?
 - From the region?
 - From the own country?
 - From global suppliers?



Factors determining the food intake after an accident



Default food consumption (IAEA, 2001)

	Far East	Near East	Africa	South America	Central America	North America	Europe	Oceania
Water (m ³ /a)	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6
Milk (L/a)	65	140	80	135	155	325	250	410
Meat (kg/a)	40	55	35	90	75	205	100	200
Grain, root crops, vegetables and fruits (kg/a)	510	600	380	470	445	535	410	500
Freshwater fish (kg/a)	35	10	15	20	25	25	30	15
Marine fish (kg/a)	60	20	30	35	45	40	50	30
Shellfish (kg/a)	20	5	10	10	15	15	15	10



IAEA, Report SRS-19, 2001



	Consumption rates (g d-1)					
Foodstuff		fo	r age gro	up		
	1 a	5 a	10 a	15 a	adults	
Spring wheat,	0.7	1.4	1.8	2.0	2.6	
whole grain						
Spring wheat, flour	3.9	8.1	10	12	15	
Winter wheat,	6.0	13	16	18	23	
whole grain						
Winter wheat, flour	35	73	91	100	130	
Rye, whole grain	2.2	4.8	6.0	6.9	8.7	
Rye, flour	9.3	19	24	28	35	
Oats	2.9	3.1	3.9	4.4	5.6	
Potatoes	45	35	60	83	160	
Leafy vegetables	58	74	79	86	94	
Root vegetables	21	24	29	33	33	
Fruit vegetables	12	36	41	46	47	
Fruit	150	72	91	100	120	
Berries	0	10	12	14	14	
Milk	560	140	180	210	230	
Condensed milk	0	11	14	16	18	
Cream	0	9.6	13	14	16	
Butter	0	6.1	9.5	12	18	
Cheese (rennet)	0	10	14	19	26	
Cheese (acid)	0	6.6	8.9	12	17	
Beef (cow)	1.5	18	19	23	27	
Beef (cattle)	3.0	35	38	46	55	
Veal	0.2	1.4	1.5	1.8	2.2	
Pork	3.9	72	78	90	108	
Chicken	1.5	11	12	14	17	
Eggs	5.0	18	25	36	43	
Beer	0	0	12	130	610	



Model Description of the Terrestrial Food Chain and Dose Module FDMT in RODOS PV6.0 RODOS(RA3)-TN(03)06

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Another example:

Age-dependent German consumption rates V_k as applied as default in FDMT



ENEP Predicted doses via different exposure pathways following the Chernobyl deposition (Southern Bavaria, Germany)

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Testing the model: Cs-137 whole body counting ENEP (near Munich)



Müller, H, and Pröhl, G., The radioecological Model ECOSYS: Concept and Applications, Proc. Intern. Workshop on Improvement of Environmental Transfer Models and Parameters, Tokyo, Japan, 5-6 February, 1996

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Improving confidence in model results

• Input data

- Uncertainty increases with number of steps between input and endpoint
- Prefer input data 'closest' to the endpoint
 - In-vivo measurements: Whole body, thyroid
 - Individual dosimeters
- Temporal and spatial resolution in of monitoring data

• Model analysis

- -Comparison with real data
- Identification of sensitive assumption and parameters
- Systematic uncertainty analysis



Monitoring and models

Monitoring

- Providing measures results
- Validate and calibrate models
- But: How representative are measurements? Do they always tell the "truth"?

Models

- Understand measurements
- Interpolation in time and space
- Extrapolation to the future
- Overcome data gaps

Decline of ¹³⁷Cs-levels in food and water



Smith, J.T. et al. (2000) Chernobyl's legacy in food and water. Nature, 405.

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Ecological half-life: 10.5 years Effective half-life (including decay): 7.8 years



¹³⁷Cs in wild boar collected in South Germany (1986 to 1999)





Time-dependence of ¹³⁷Cs-TFs for brown and white rice in Japan

S. Fesenko, N. Sanzharova, K. Tagami: Evolution of plant contamination with time (IAEA-TECDOC 1616), 2009)



Summary of ecological half-lives for Cs

Plants and animal food products on agricultural land - 4 to 6 years

Pasture

- In the first 5-6 a after deposition:
 - **1 4 years**
- Afterwards: **5 15 years**
- Slower decline for vegetation on <u>peat</u> and mineral soils <u>low in clay</u> minerals.

Forest products (Roe deer, deer, wild boar, forest plants, berries, fungi (Middle Europe))

- Average: 12 years
- In some areas much longer