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Assessing potential exposures to people in the post-closure period of a waste disposal facility

III. Option A: Elaborating an envelope for future conditions – The analogue approach

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Example for option A: Analogue approach

The site considered

Disclaimer

- The site considered does not refer to a site where a waste disposal facility is planned.
- The data have been taken from studies to explore the possible feasibility of a waste disposal facility at this location.
- The activities to study the site terminated in the early 2000s.

General characteristics of the area

- The site is located in Northern Germany.
- The distance to the sea is about 100 km.
- For estimating possible impacts a region is considered in the vicinity of the site with an area of about 100 km².
  - The altitude above sea level is 17-20 m.
  - Topography is flat with subdued relief.

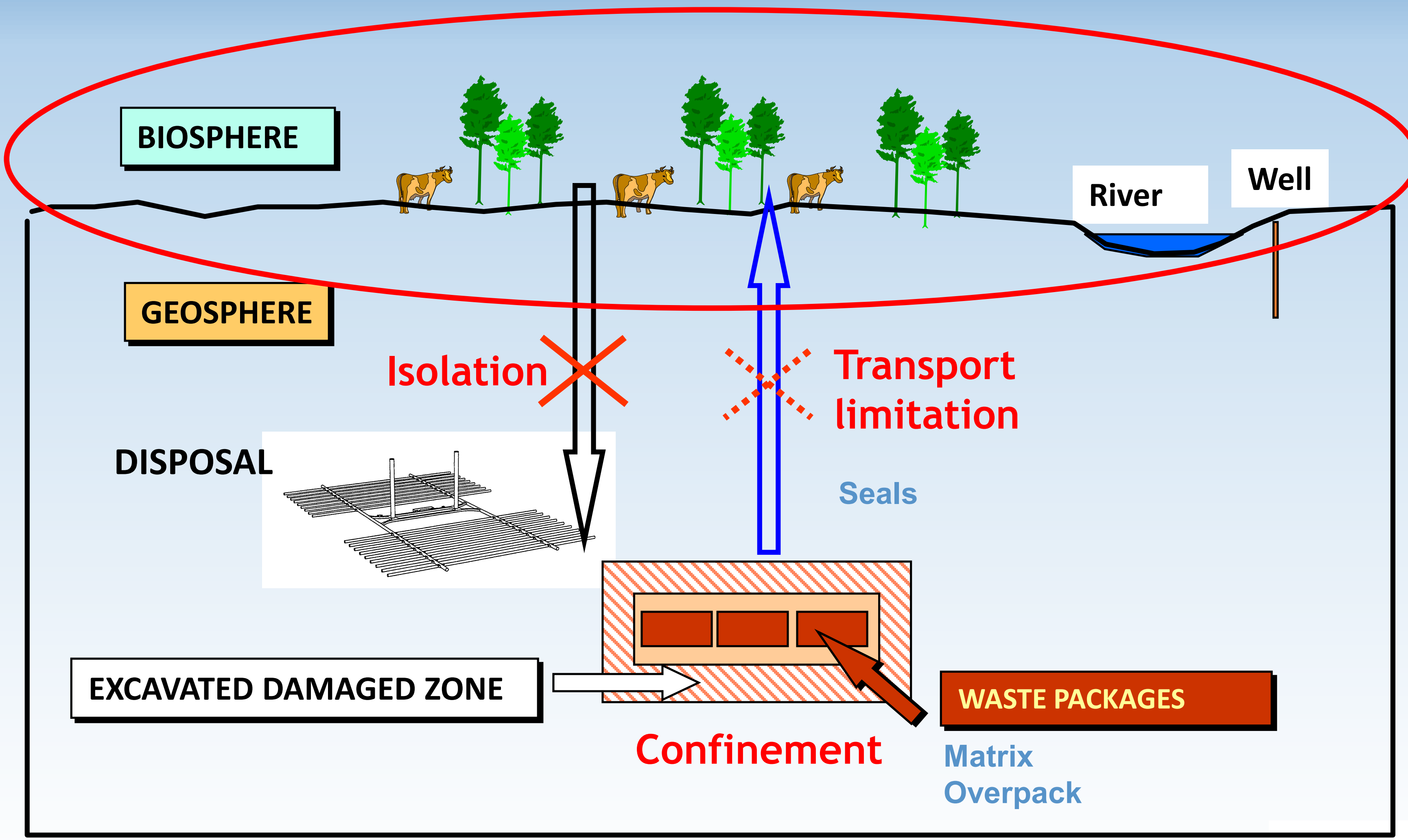
Soil types

Soil type	Origin/horizons	Height of water table	Irrigation
Podsol-gley	**Organic soils** developed on fens, that dried out due to a decreasing groundwater level 0-38 cm: fine sand (2-4% organic matter O.M.)	3-10 m	**Yes**
Podsol (most important)	**Wind borne sand** over fluvial sediments 0-30 cm: fine sand (4-8 % O.M.)	1-2 m	**Yes**
Gley	**Gley** on fluvial sediments 0-30 cm: plough horizon (clayey silt) (2-4 % O.M.)	ca. 0.6-1.6 m	**No**

Present climate

- **Reference station**
  - Magdeburg (Germany)
- **Climate classification (Köppen)**
  - Temperate, Cfb
  - Relatively cool summers
  - Mild winters
- **Mean annual temperature**
  - 8.8°C
- **Mean precipitation**
  - 520 mm/year

IAEA Specific Safety Guide 14 (SSG-14)
Geological Disposal Facilities for Radioactive Waste



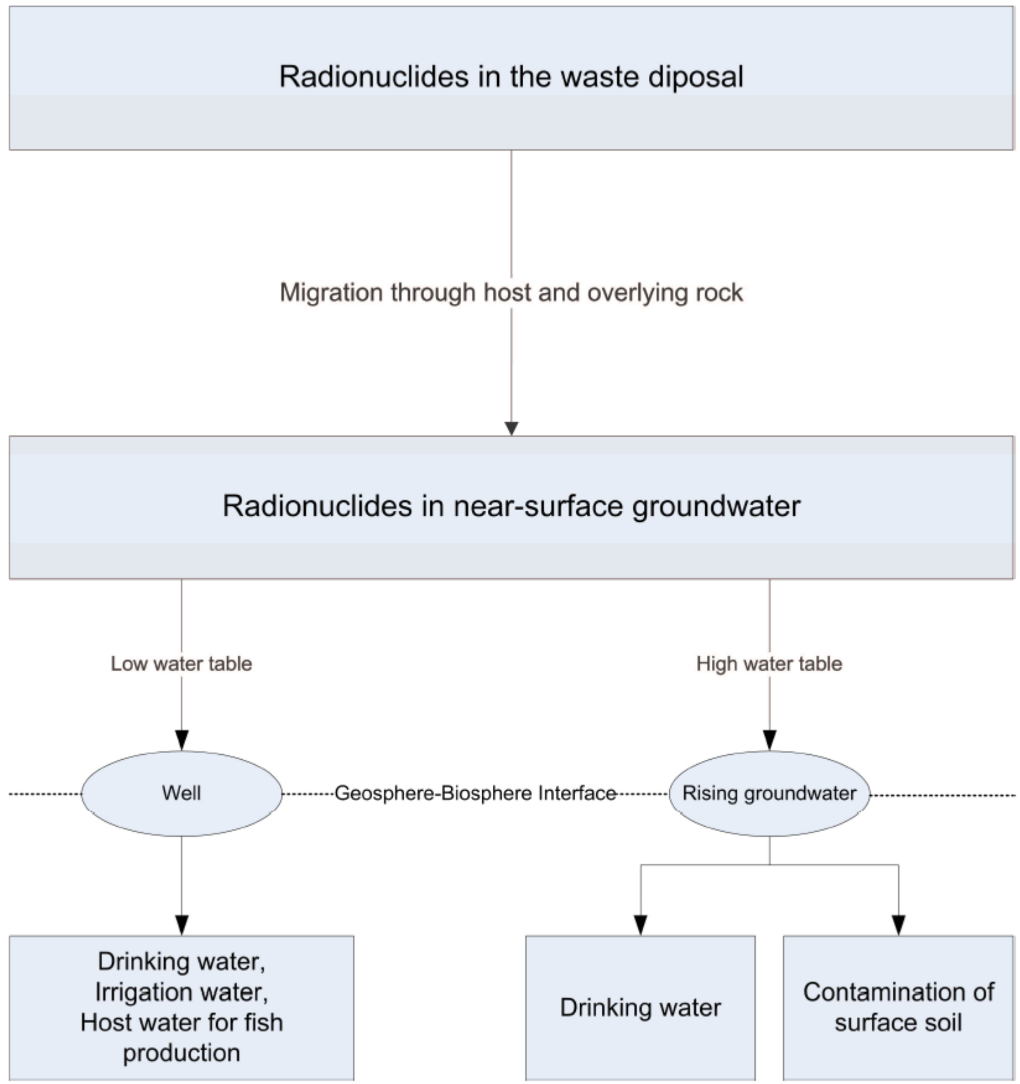
Radionuclides enter the biosphere

From the disposal area to the near-surface aquifer

- Radionuclides are released from the disposal
- Radionuclides migrate through the overlying rock
- Radionuclide contaminate the near-surface aquifer

From the near-surface aquifer to the biosphere

- Withdrawal of water via a well
- In case of high ground-water level, radionuclides may directly contaminate soils



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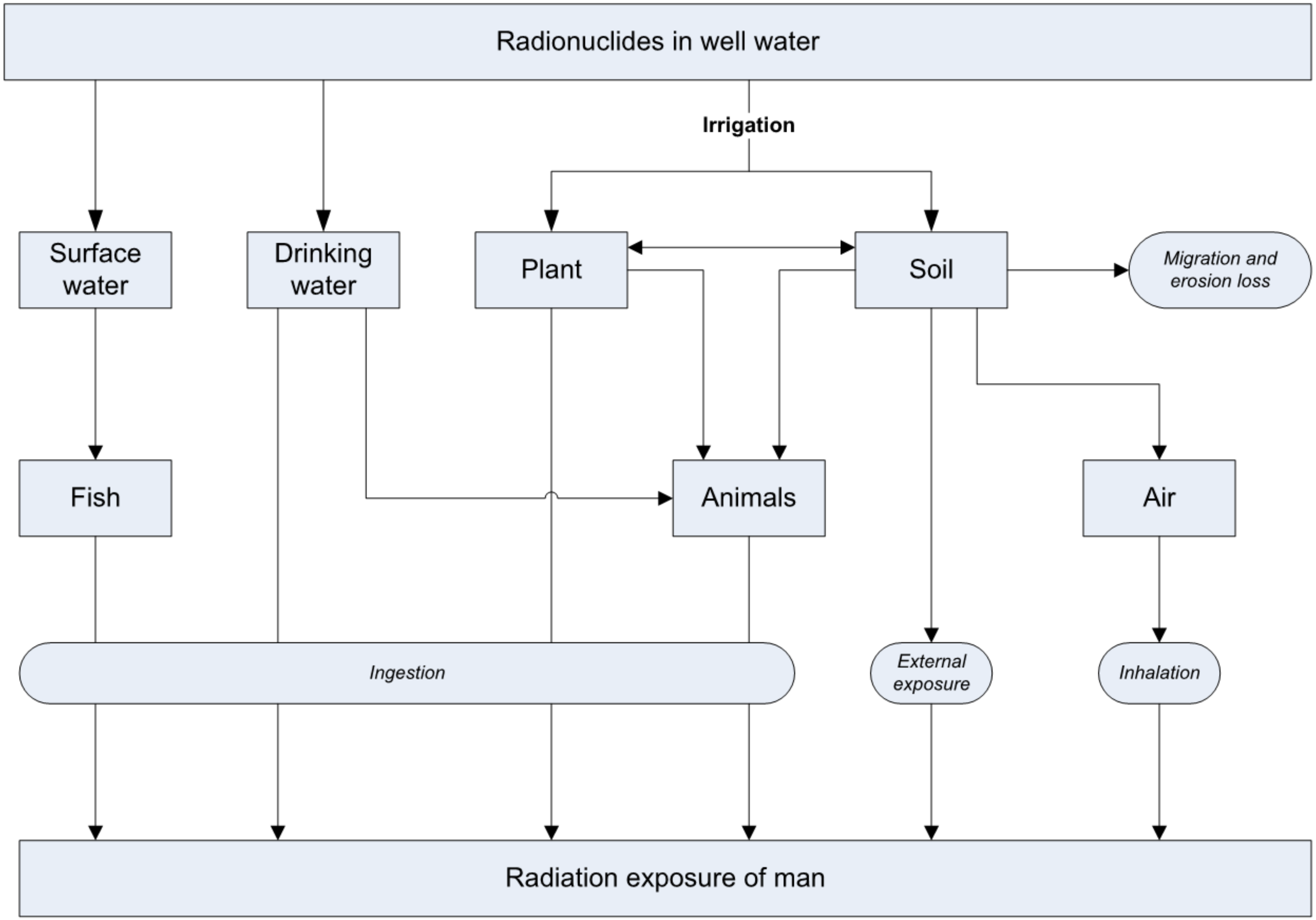
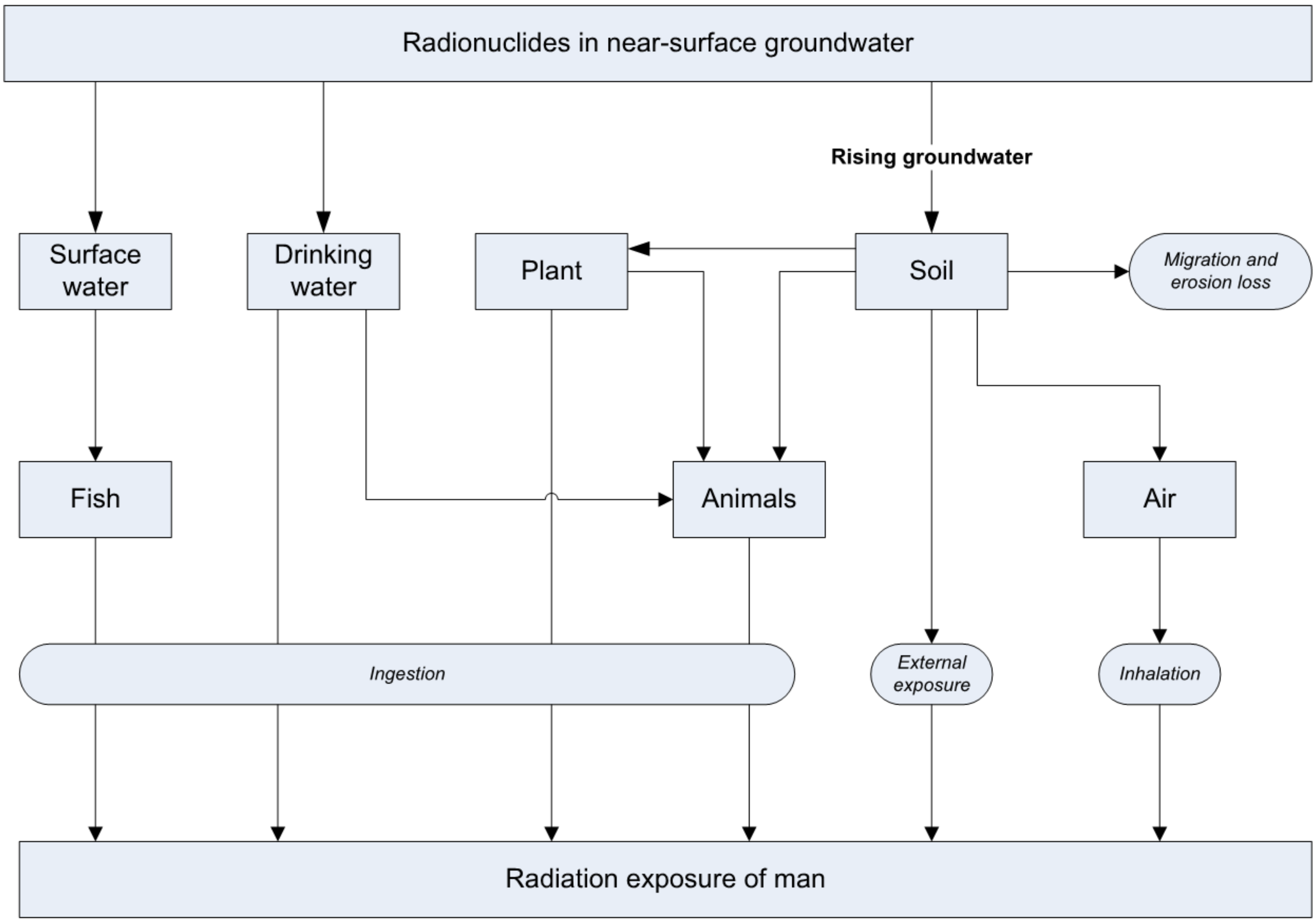
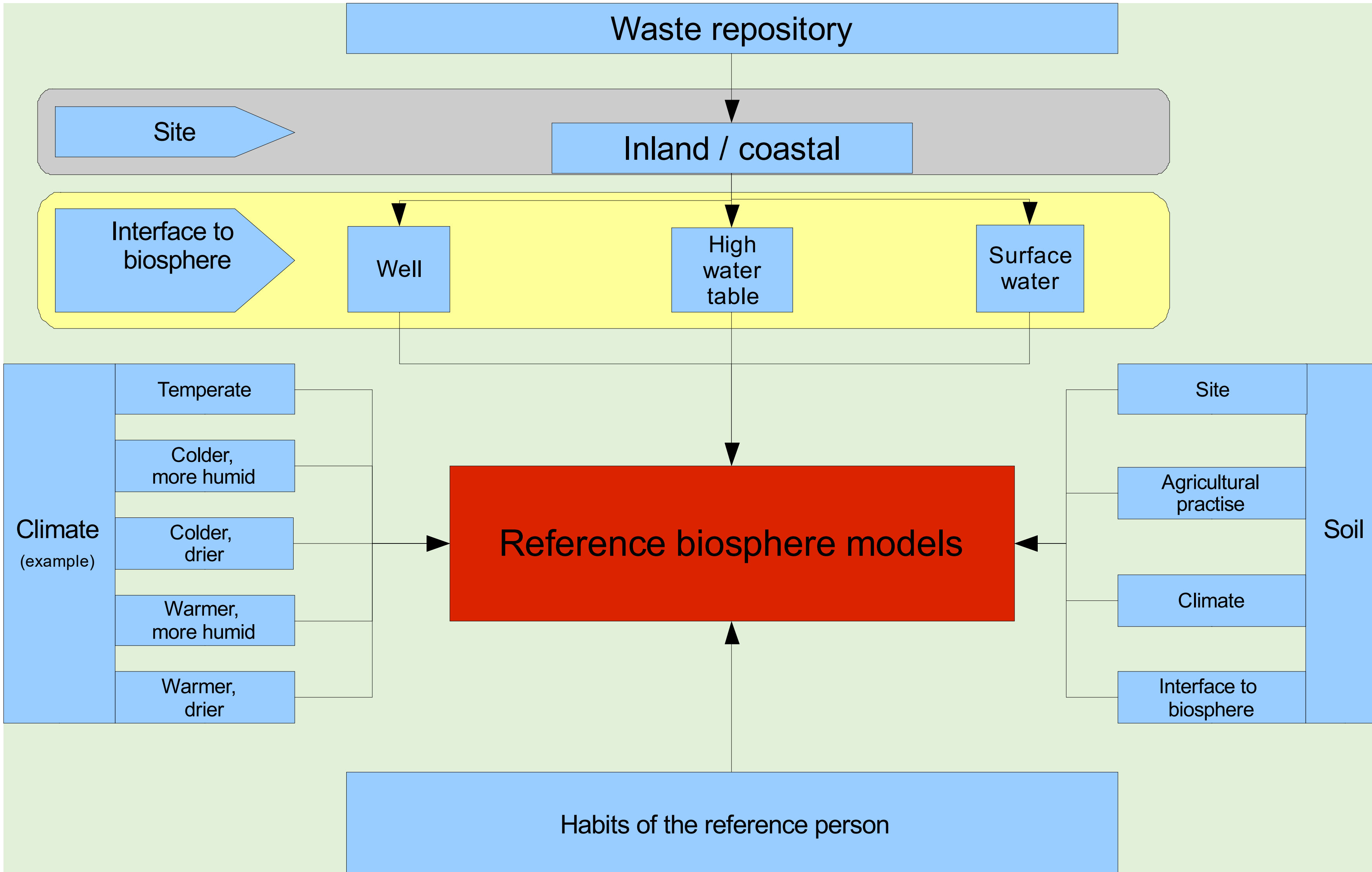
Pathways for withdrawal of water from a well



Pathways for rising ground water

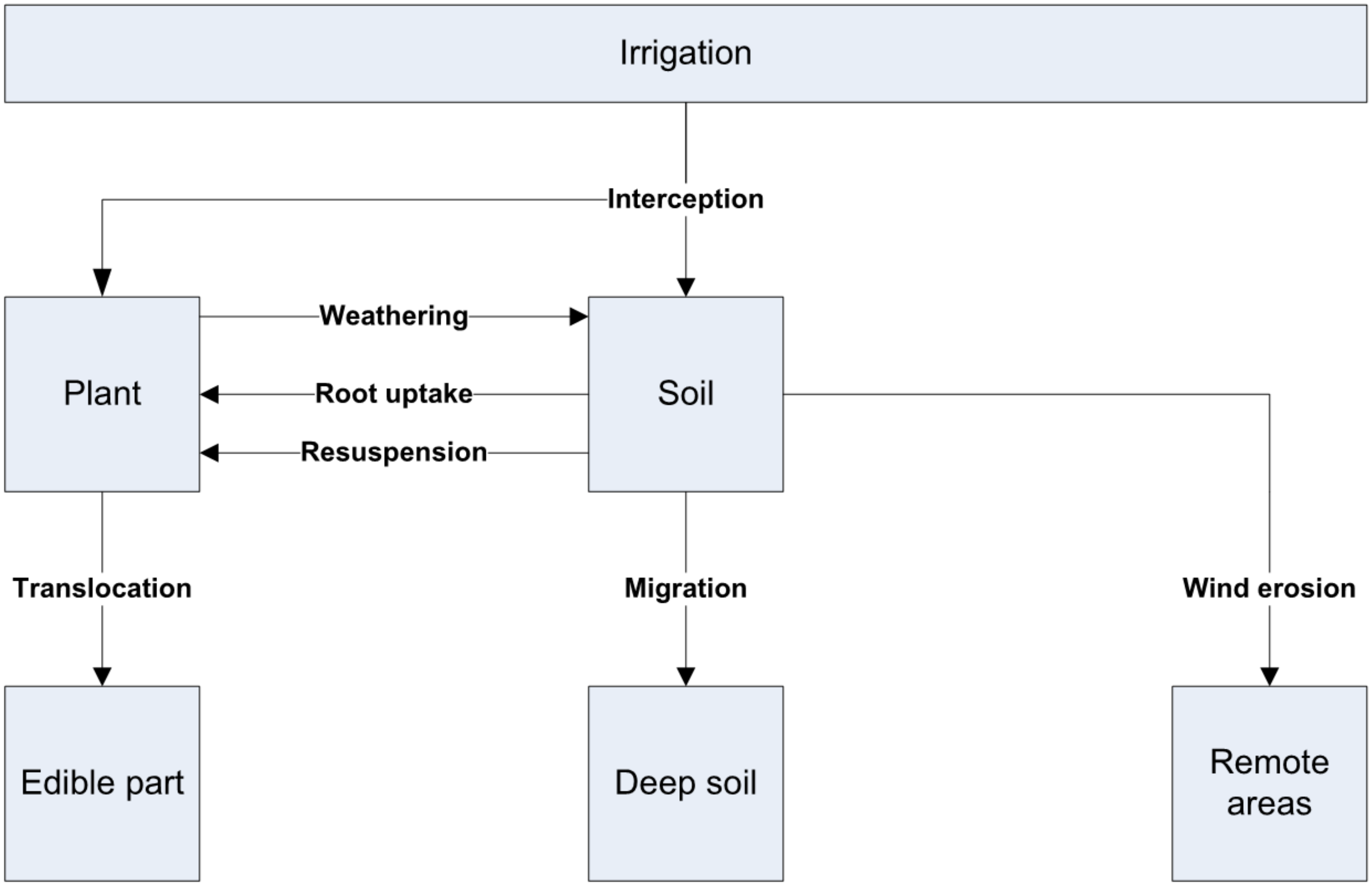


Addressing future developments

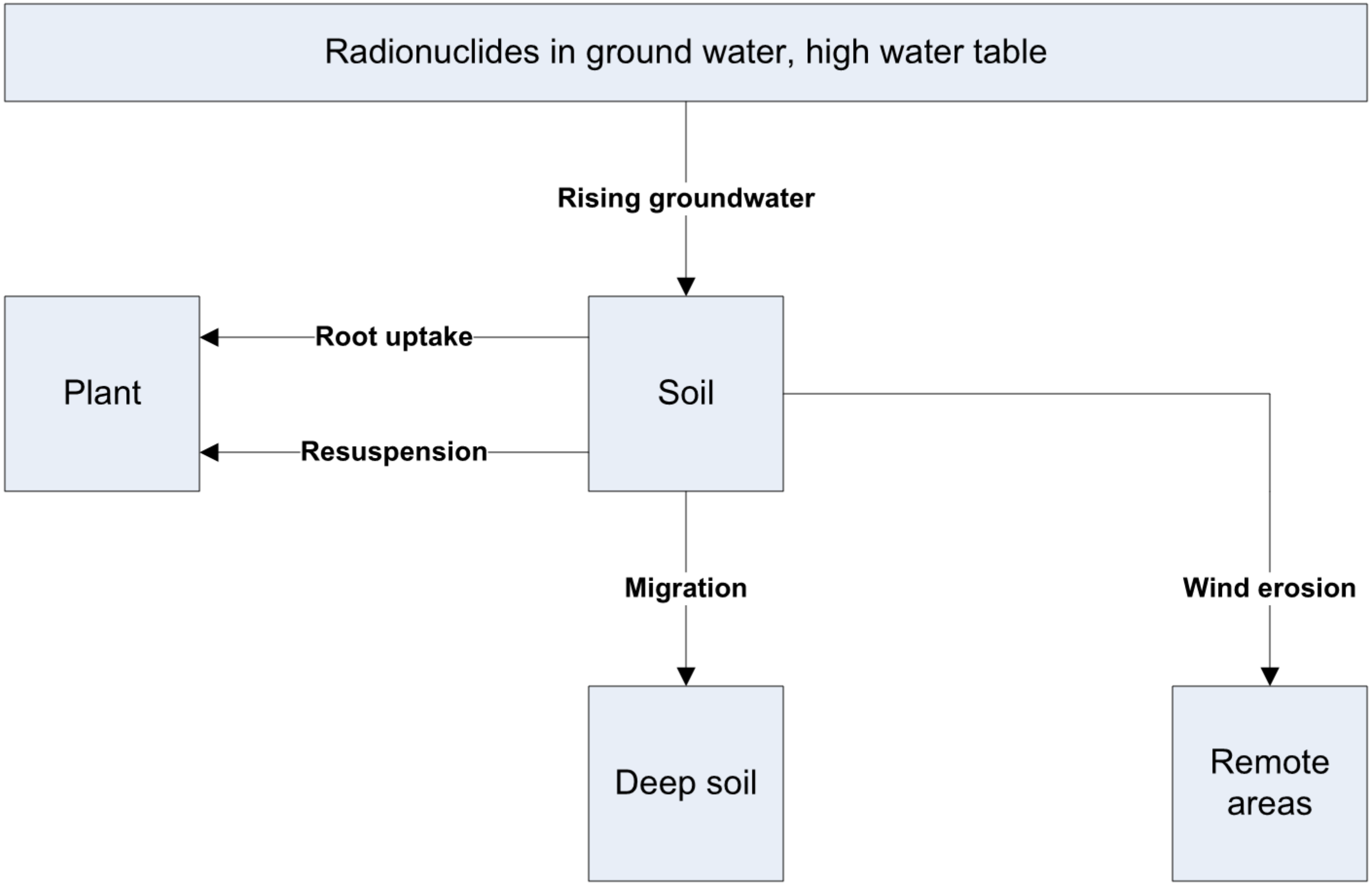


Transfer processes involved

Plant model irrigation



Plant model for rising groundwater



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Relevant processes

Contamination process	Geosphere-biosphere interface	
	Well	Rising groundwater
Transfer to plants		
Radionuclide uptake from soil	X	X
Contamination due to resuspension	X	X
Weathering	X	
Interception by plants during application of irrigation water	X	
Translocation (systemic transport within the plant subsequent to foliar deposition)	X	
Loss from soil		
Migration	X	X
Erosion	X	X
Transfer to animals		
Drinking water for cattle	X	X
Use of contaminated feed plants	X	X
Contamination of air by resuspension		
Accumulation of radionuclides in the resuspendable soil fraction	X	X
Transfer to freshwater fish		
Radionuclide uptake by fish	X	X
Attachment of radionuclides to particles and sedimentation	X	X

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Dependency of processes

Process	Universal process	Climate	Soil	Technology
Transfer to plants				
Amount of irrigation water		**xxx**	x	
Radionuclide uptake from soil			**xx**	x
Contamination due to resuspension		**xxx**	**xxx**	
Weathering	xxx	x		
Interception by plants during application of irrigation water	xxx			
Translocation (systemic transport within the plant)	xxx			
Loss from soil				
Migration		**xx**	**xx**	
Erosion		**xx**	**xx**	xx
Transfer to animals				
Drinking water for cattle		**xx**		x
Use of contaminated feed plants				x
Contamination of air by resuspension				
Accumulation of radionuclides in the resuspendable soil fraction			**xxx**	
Transfer to freshwater fish				
Radionuclide uptake by fish	xxx			
Attachment of radionuclides to particles and sedimentation	x			
Living habits				
Food and water intake		**xx**		x
Time of staying out doors		x		x

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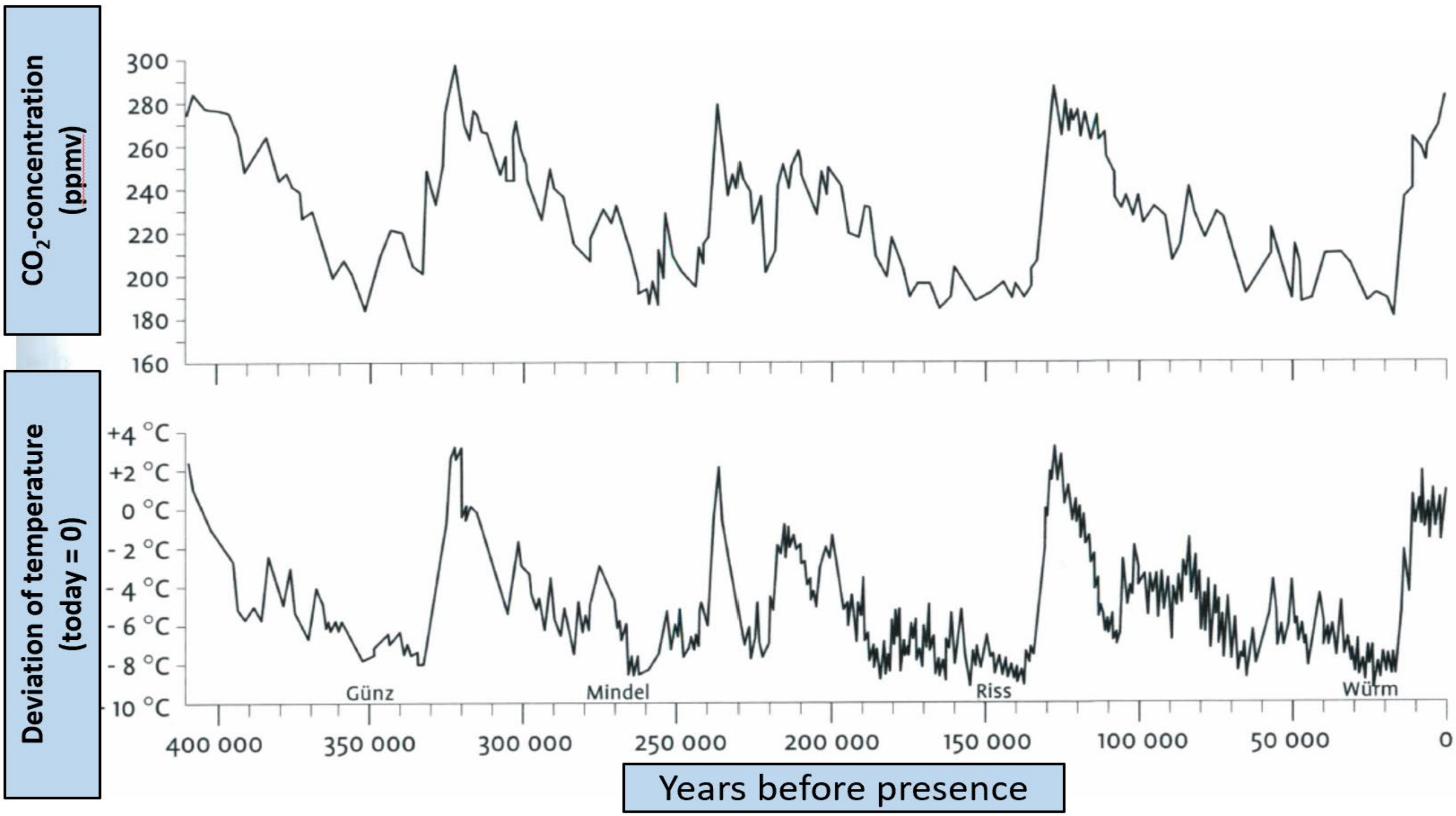
Climate and irrigation

Temperature and CO_2 in the last 400 000 a



Maximum temperature variation: 12 K

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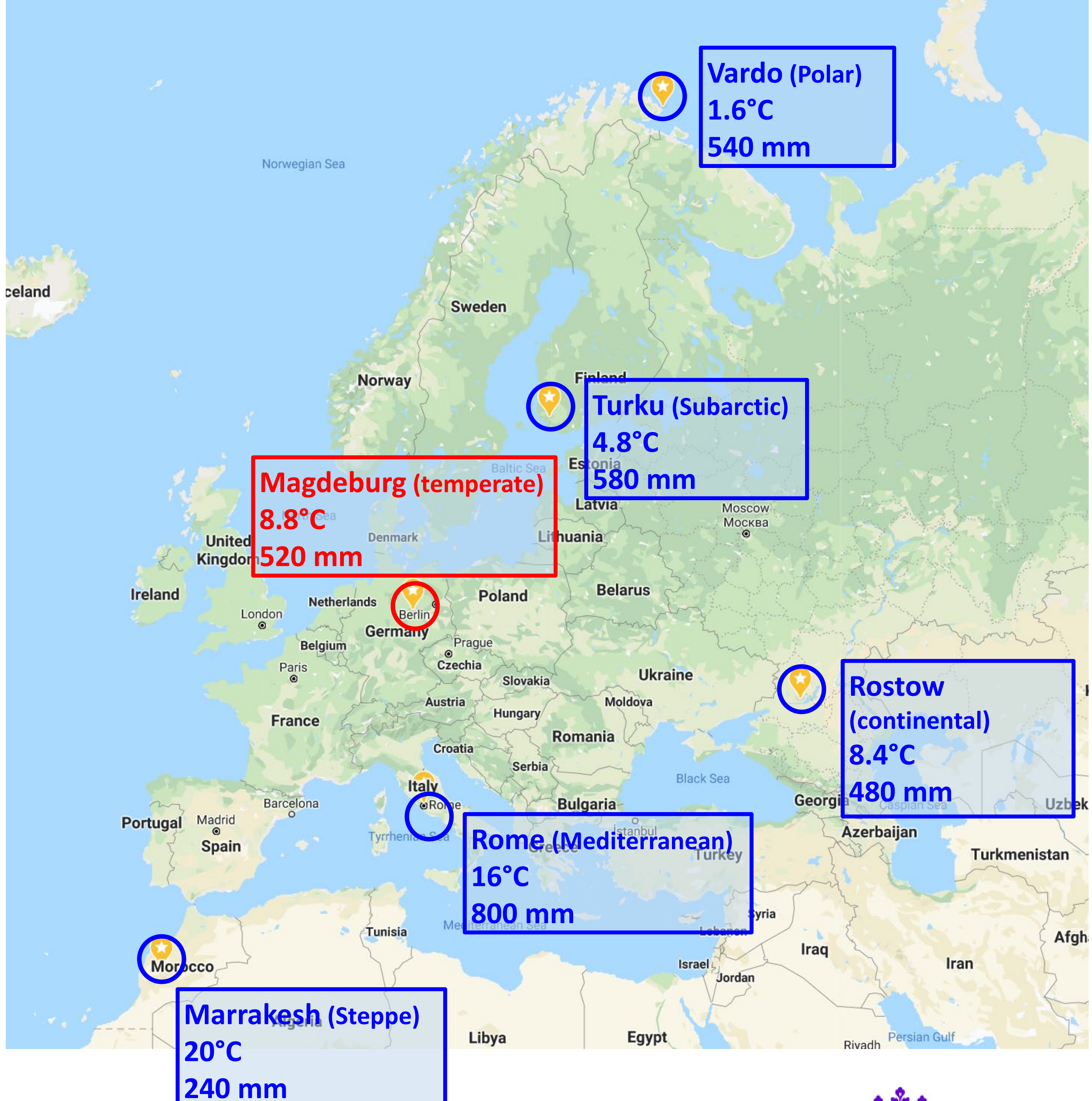
Identification of locations in Europe that reflect variations of climate in the last 400000 years:

The Site and **analogue sites** considered to reflect climate change



Variation of mean temperature: 1.6 -20 K, Δ=18.4 K

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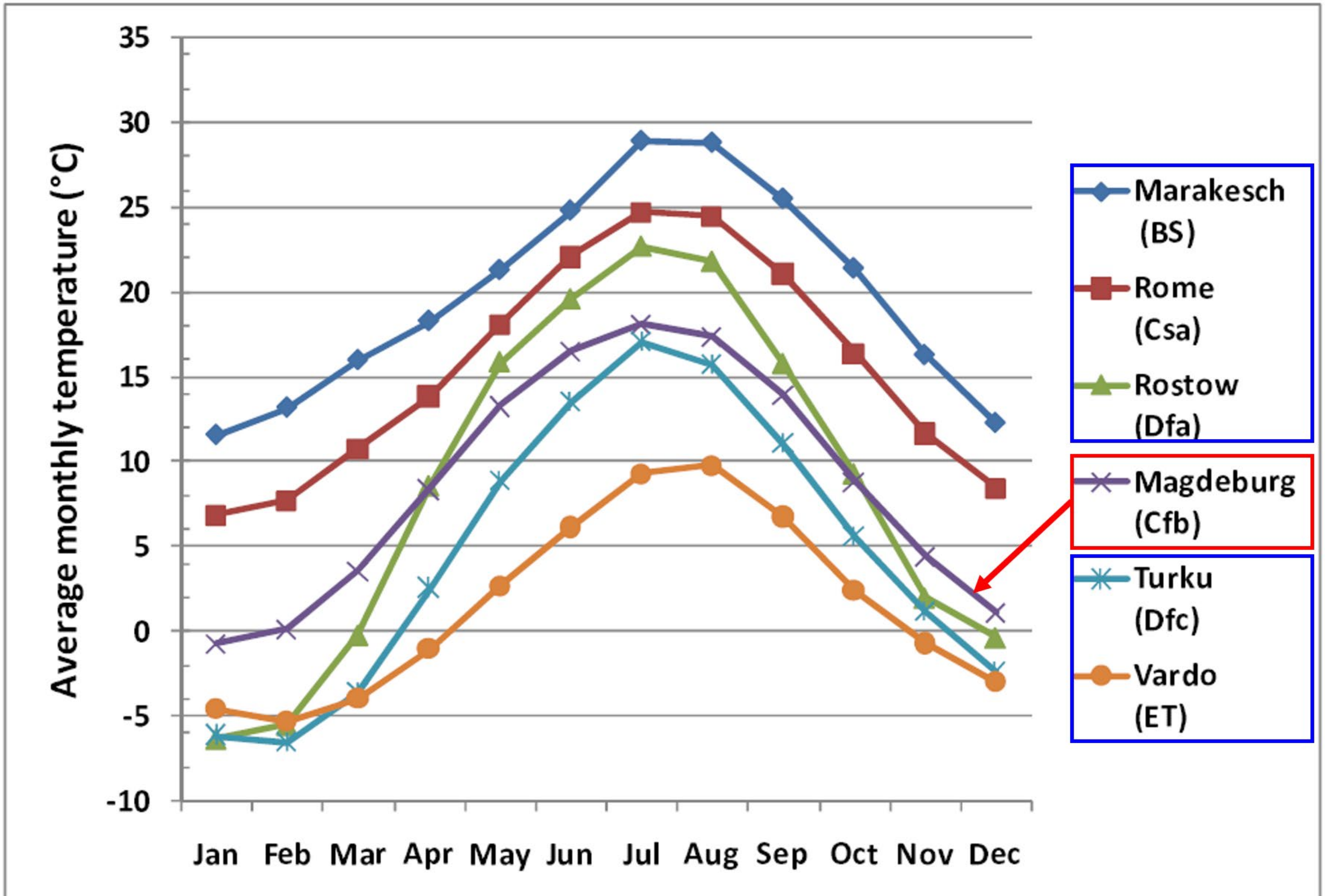
Climate and scenarios considered

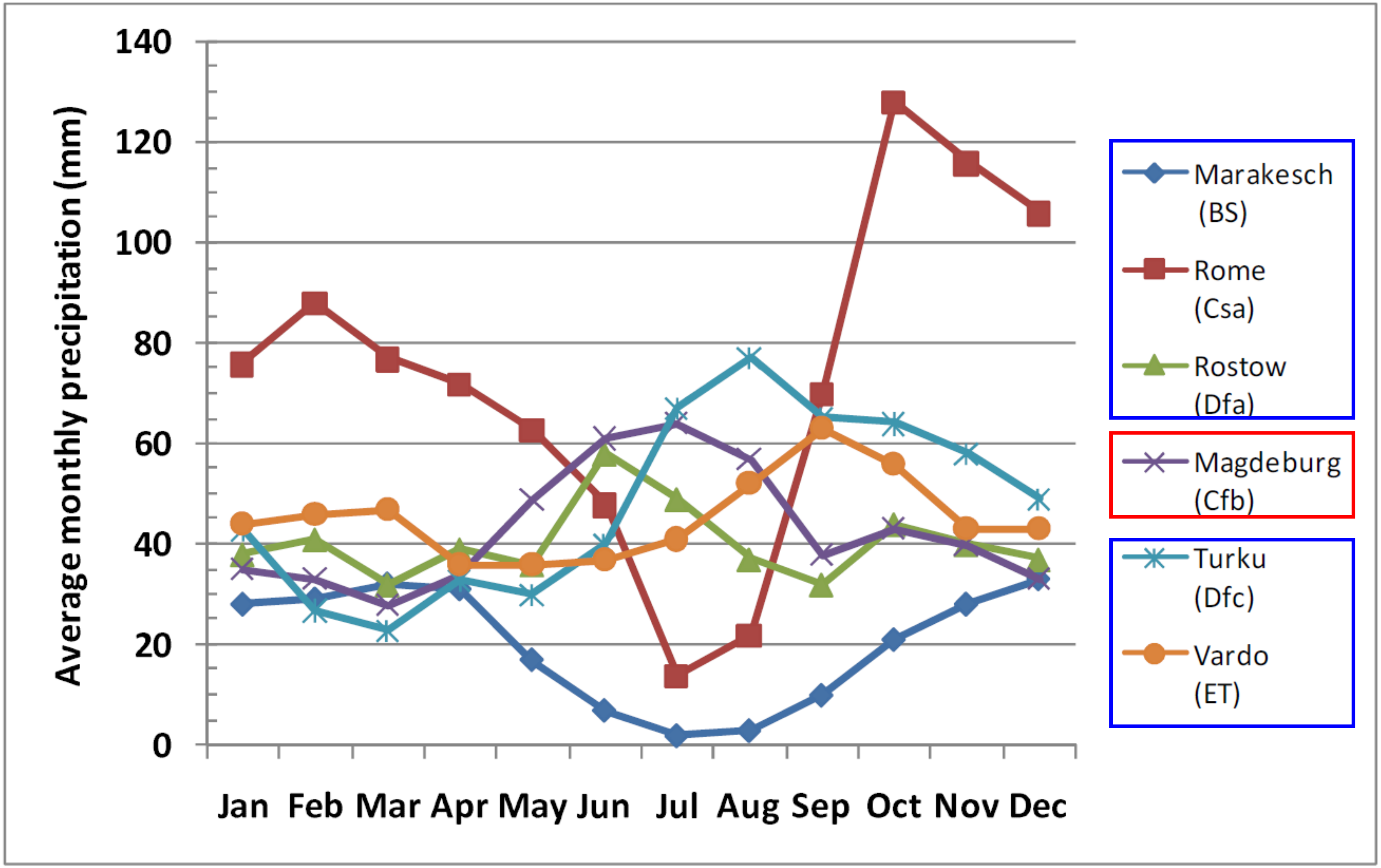
Site	Features	General characteristics	Mean tempe-rature	Mean precipi-tation	Well	Rising ground-water
Present day climate at the site considered						
Magdeburg Cfb	Temperate	Cool summers Mild winters	8.8	520	x	x
Analogue climates as an envelope for future developments						
Rome Csa	Mediterranean hot summer climate	Hot summers Humid winters	15.5	800	x	-
Marrakesh BS	Steppe (semi-arid)	Hot summers	19.9	240	x	-
Rostow Dfa	Hot summer continental climate	Hot summers Cold winter	8.4	480	x	x
Turku Dfc	Subarctic (Boreal)	Cold summer Very cold winters	4.8	580	x	x
Vardo ET	Polar (Tundra)	Cold summer Very cold winters	1.6	540	x	x

Variation of mean temperature: 1.6 -19.9 K, Δ=18.3 K

Average monthly temperatures at the site and the analogue sites



Average monthly *precipitation* at the site and the analogue sites



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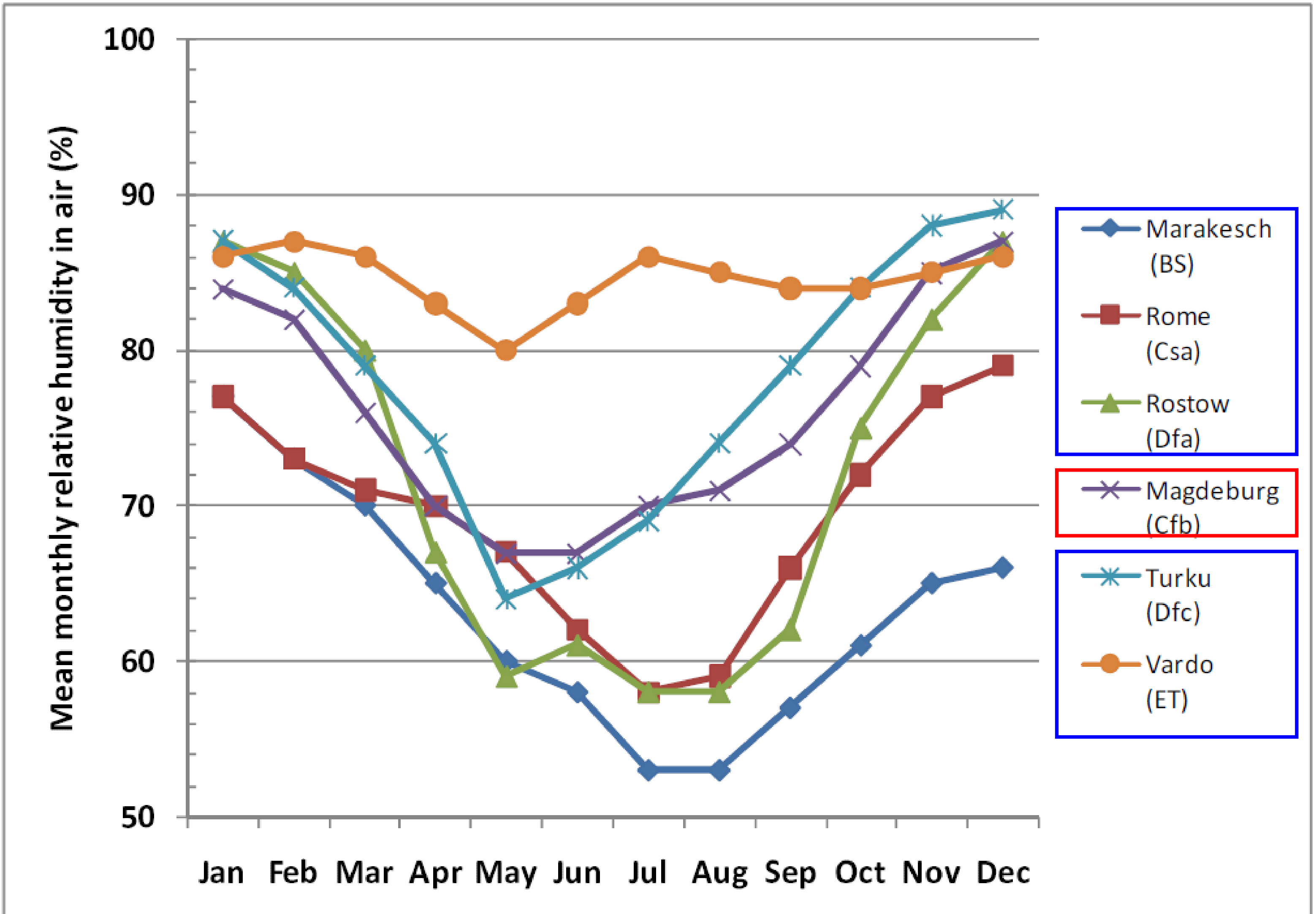
Average monthly *relative humidity* at the site and the analogue sites



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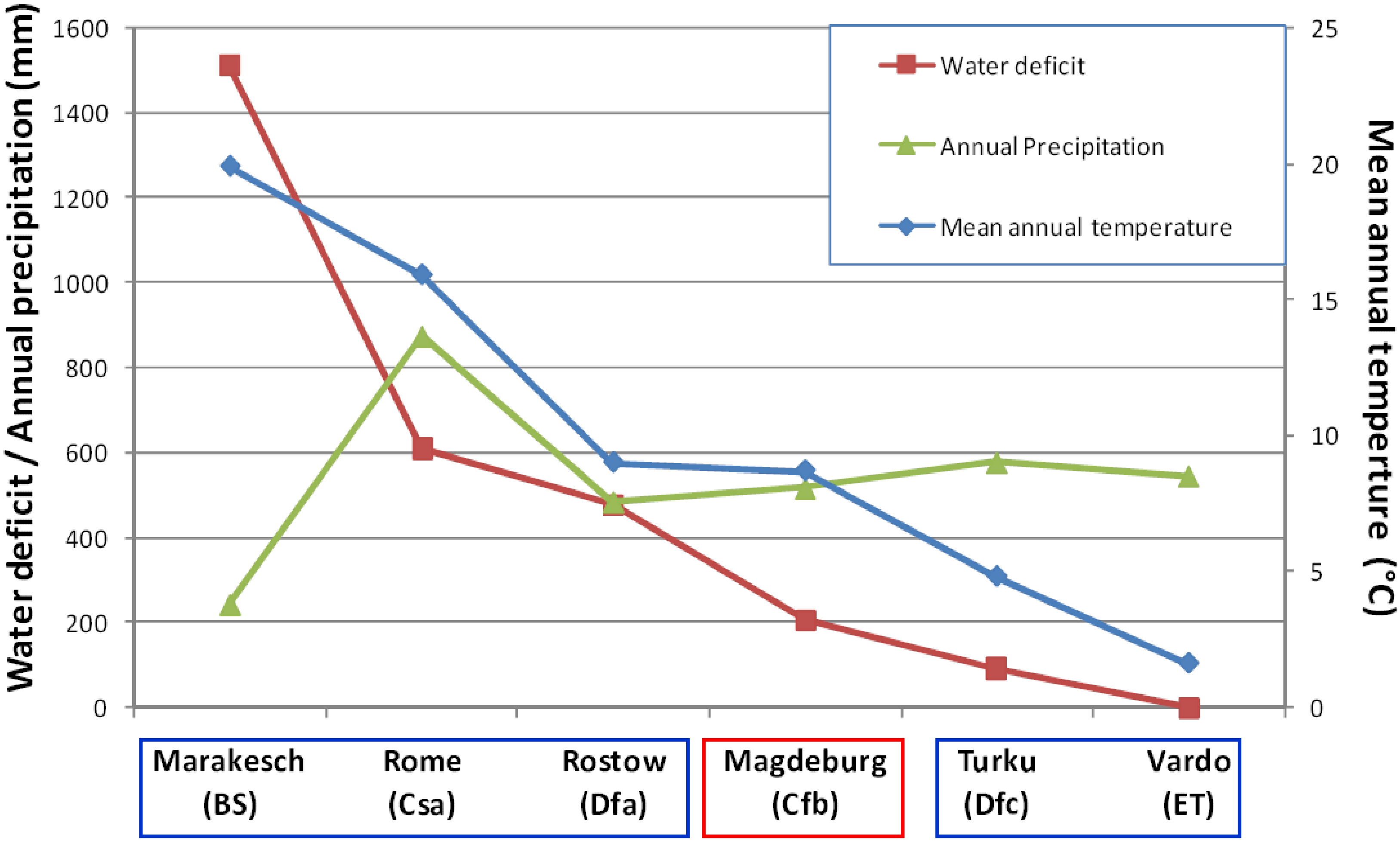
Irrigation

- **A key process for radionuclides for entering the biosphere**
- **Dependence on soil and climatic conditions**

- **The water storage capacity of soil**s declines in the order clay > loam > sand
  - In **clay** soils, water is relatively strongly bound.
  - **Loam** soils provide the best conditions for water storage
  - In **sandy** soils, their water storage capacity is lowest.

- **Temperature, precipitation and relative humidity** determine the water deficit of a site
  - With **increasing temperature**, more water is lost by evaporation and transpiration of plants.
  - With **increasing relative humidity** the evapotranspiration and, subsequently, the water deficit decrease.

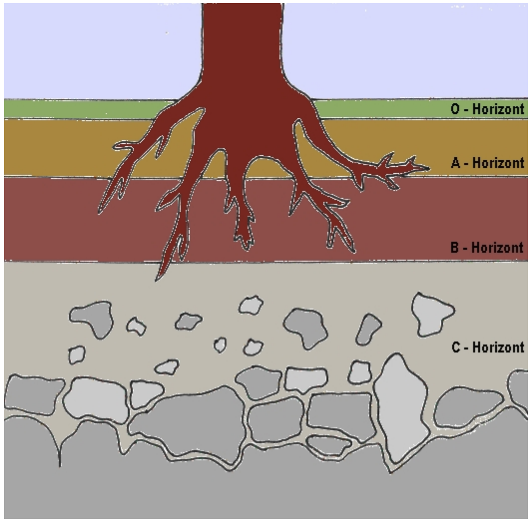
Precipitation, temperature und *water deficit* at the site and the analogue sites



Typical irrigation regimes (l/m² a)					
Marrakesh Morocco	Magdeburg, D	**Rome, Italy**	Rostow, Russia	Turku, Finland	Vardo, Norway
300-700	***100-200***	***350-600***	***400-500***	***0-100***	***0***

Irrigation needs

Aspects of long-term behaviour of radionuclides in soil



Speciation und mobility

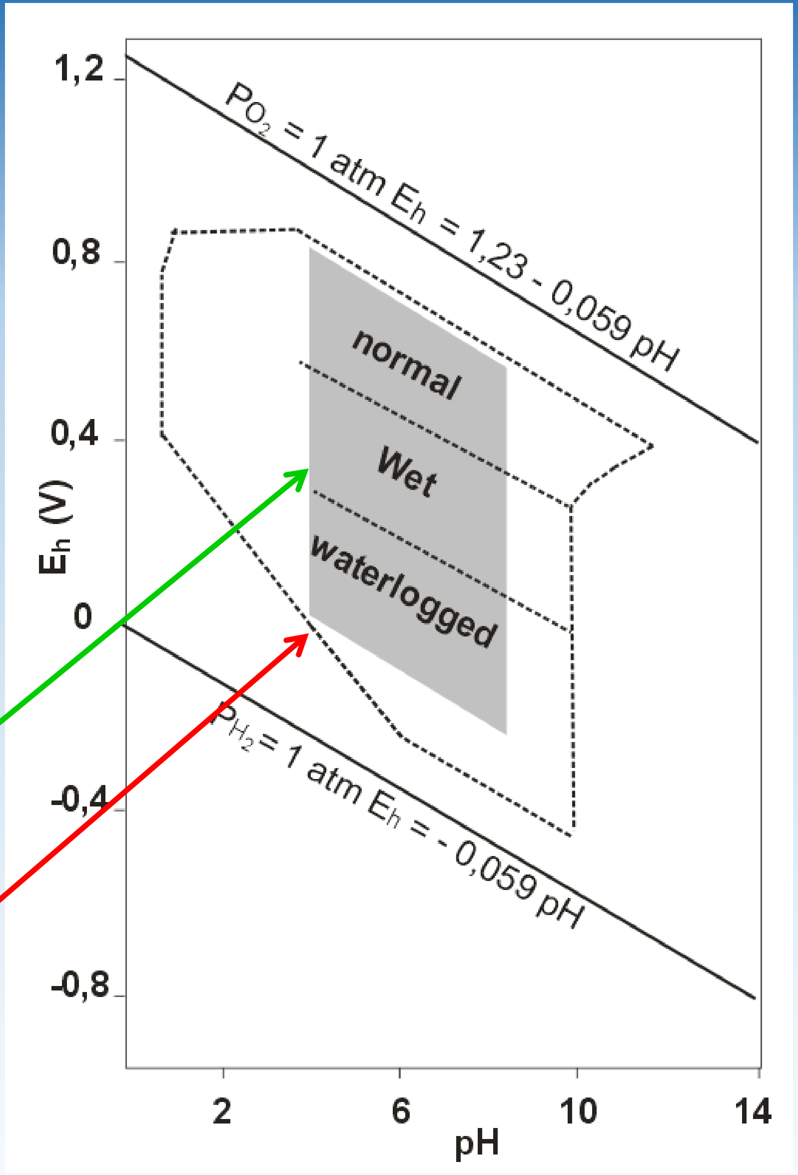
- **Sorption capacity**
  - Clay, silt and sand
  - Organic matter
- **pH-value**
- **Redox potential**
  - Quantifies the oxygen status in soil
- **Soil management aims to achieve the following conditions to optimize growth**
  - Organic matter between 2-4 %
  - pH between 5.5 and 7.5
  - Redox potential between 100 and 600 mV
  - Optimize water supply
  - Enable soil aeration
  - Provide nutrients

Redox, pH and soil water:

=> **Key factors for the radionuclide availability in soil**

Limit for successful agriculture

Limit for micro-organsims in soil



Chemical reactions in dependence on redox

Status of soil aeration	Redoxreaktion	Redoxpotential E_{h7} (mV)
Well aerated soils	Start of NO_3^- - reduction Start of Fe^{2+}	450-550 350-450
Wet soils	O_2 not detectable NO_3^- not detectable Start of formation of Fe^{2+}	330 220 150
Water-logged soils	Reduction of SO_4^{2-}, Production of S^{2-} Start of the methane production Sulphate no longer detectable	-50 -120 -180

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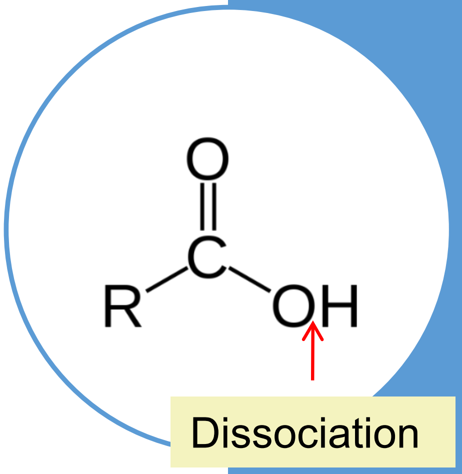
Sorption to inorganic soil components

- **Clay > silt > sand**
  - Very little dependence on pH
- **Temperate zone**
  - **Young soils (~10 000 years) => low degree of weathering of clay minerals**
  - Clay minerals are negatively charged
  - **=> Sorption of cations**
- **Tropical soils**
  - **Old soils (~1 000 000 years)** => extensive weathering of clay minerals
  - Depletion of silicates
  - Positively charged Al- and Fe-oxides remain
  - Cation exchange capacity is very low
  - **=> Sorption of anions**

Organic matter

- **Function**
  - Storage of water
  - Loosening the soil => improves aeration
  - Sorption of nutrients
- **Composition**
  - Decomposed plant and animal material, very heterogeneous
    - *Humus substances, humus acids:* persistent organic compounds with a high molecular weight
    - *Fulvic acids:* mobile organic compounds, low molecular weight, complexing agent
- **Sorption of cations to carboxyl groups**
  - Strength comparable to acetic acid (pKs: ~ 4-5)
  - Sorption decreases with pH



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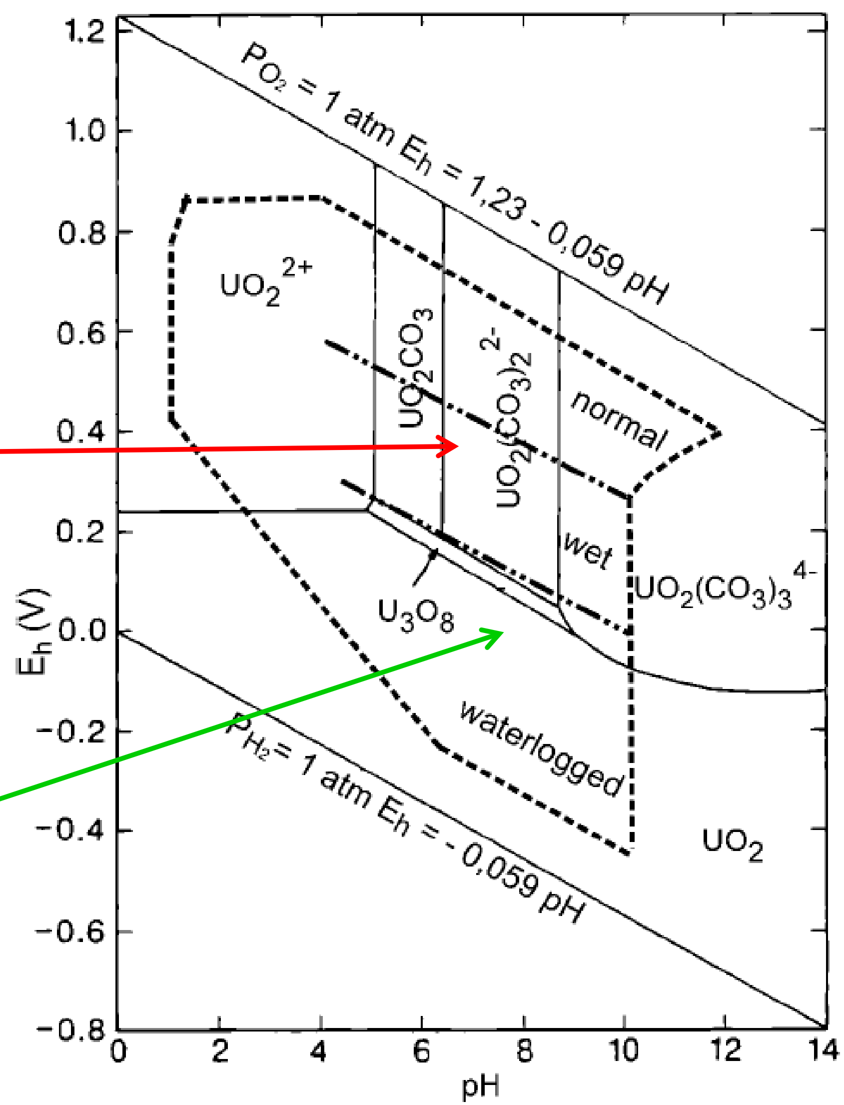
Speciation of uranium in soil

U(VI+), complexed by carbonates

U(IV+), immobile, little complexation



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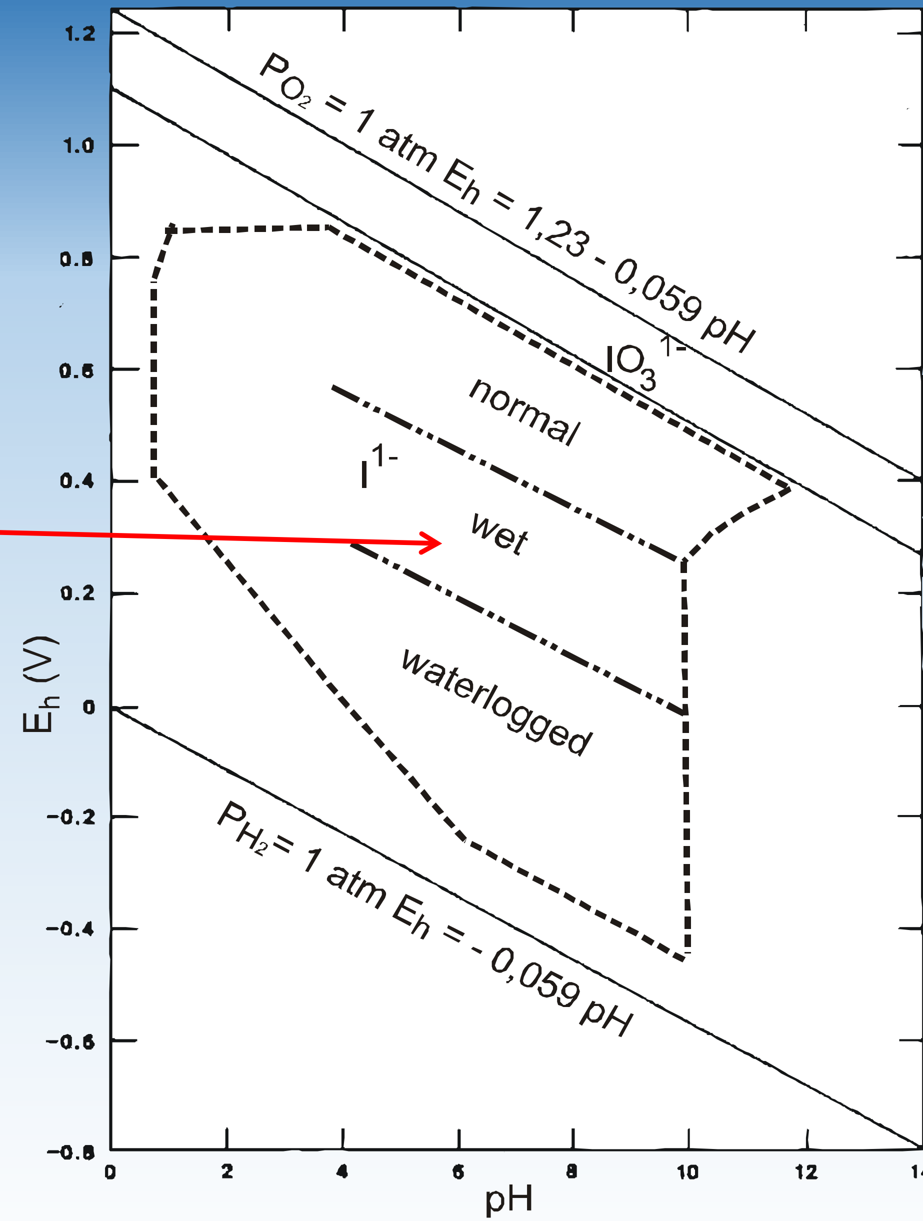
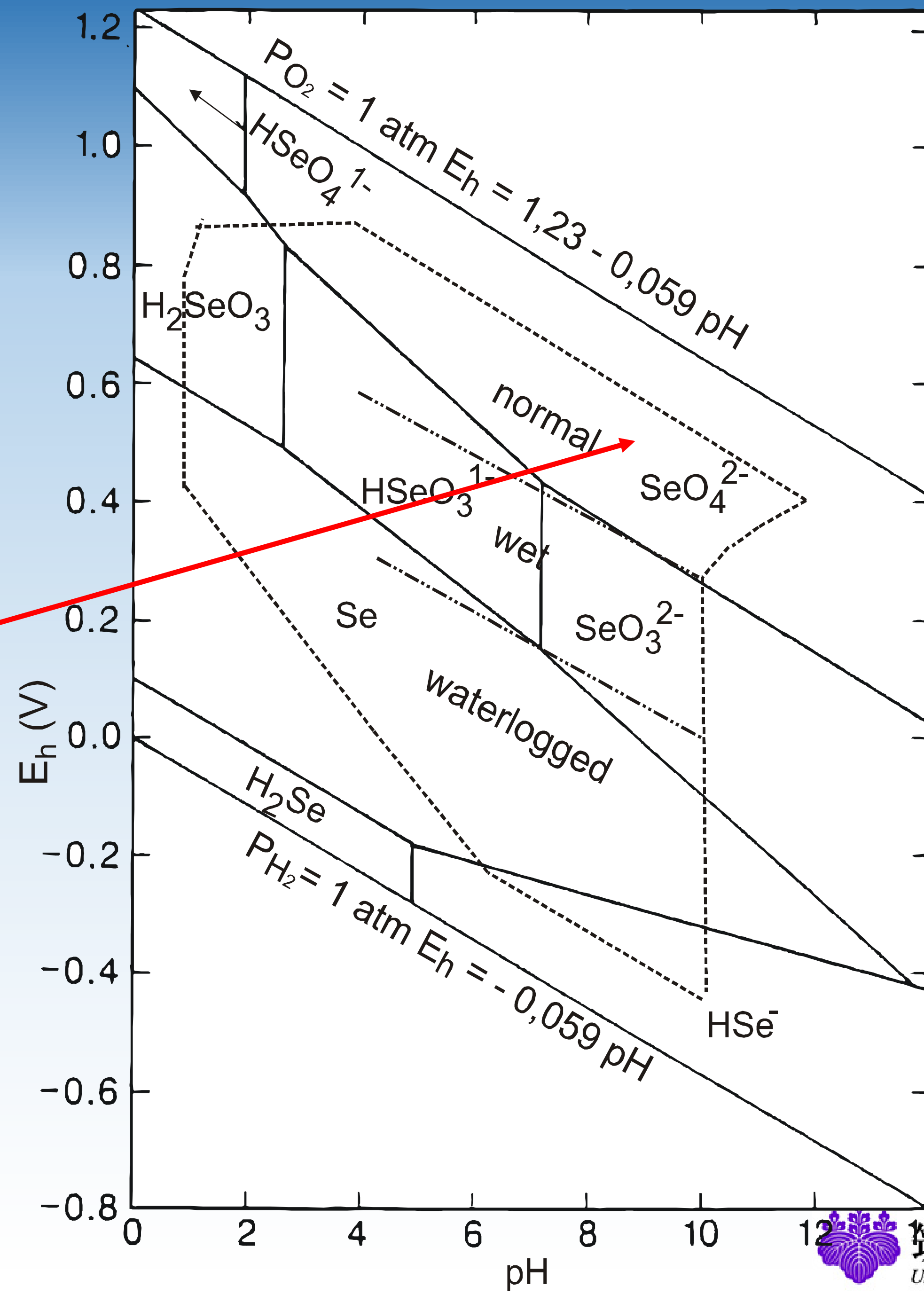
Speciation of *iodine* in soil

Available iodide present in a wide range of soil conditions



Speciation of selenium in soil

Se(VI+), high mobility



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Interaction of climate and availability in soil

- **Se-79, Tc-99, U-238 and Np-237 are most available in dry, well aerated soil**
  - For wormer climates, bioavailability tends to increase
- **I-129 is most available in wet organic soils**
  - For wormer climates, bioavailability tends to decrease
- **Cl-36 is available over a wide range of soil conditions**
  - Little influence of climate on bioavailability

Implications for long-term safety assessments

- **Scenario:**
  Withdrawal of water from a well and irrigation of crops
  - Irrigation of soils with a redox potential persistently lower than 100 mV is unlikely
  - Such soils are either water-logged or have severe structural problems
  - The optimal redox potential is near the maximum that can be achieved under normal atmospheric conditions
  - This is also valid for any future conditions.

- **Scenario:**
  Contamination of soil due to rising groundwater
  - This scenario tends to coincide with wet or water-logged soil
  - Bioavailability of Se-79, Tc-99, U-238, and Np-237 is lower as under dry conditions
  - I-129 tends to be more available

Transfer factor soil-plant and migration rates for the site considered

Radionuclide	Transfer factors soil-plant TF_{rj} (Bq/kg fresh mass plant per Bq kg^{-1} dry mass soil)						Half-life in soil (a)	
	Grass	Maize	Cereals	Potatoes & roots	Leafy veget.	Fruit veget.	Arable land (25 cm)	Pasture (10 cm)
Cl-36	**2**	**2**	**2**	**2**	**2**	**2**	**1**	**0.5**
Ni-59	0.01	0.05	0.05	5E-3	5E-3	5E-3	100	40
Se-79	**0.05**	**0.02**	**0.02**	**3E-3**	**3E-3**	**3E-3**	**50**	**20**
Zr-93	2E-3	5E-4	1E-3	1E-4	4E-4	4E-4	100	40
Nb-94	4E-3	4E-3	4E-3	1E-3	2E-3	5E-4	100	40
Tc-99	**1**	**0.1**	**0.1**	**0.1**	**1**	**0.3**	**5**	**2**
Pd-107	0.03	0.03	0.03	5E-3	0.02	5E-3	100	40
Sn-126	5E-3	5E-3	5E-3	1E-03	3E-3	1E-3	100	40
I-129	**0.1**	**0.01**	**0.01**	**0.01**	**0.01**	**0.01**	**100**	**40**
Cs-135	0.05	0.02	0.02	0.05	0.05	0.02	100	40
Ra-226	0.02	1E-3	1E-3	0.01	0.01	0.01	100	40
Th-230	0.01	5E-4	2E-3	2E-4	1E-4	2E-4	200	80
Pa-231	5E-4	2E-4	2E-4	1E-4	3E-4	1E-4	200	80
U-238	2E-3	2E-3	2E-3	1E-3	5E-3	1E-3	200	80
Np-237	**0.01**	**3E-3**	**3E-3**	**2E-3**	**2E-3**	**2E-3**	**100**	**40**
Am-243	2E-4	2E-5	2E-5	1E-4	1E-4	1E-4	200	80
Pu-239	1E-4	1E-5	1E-5	1E-5	1E-4	1E-5	200	80

Radionuclides, which are typically most important in long-term safety assessments

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Based on considerations of water status, redox and pH
Modification of transfer factors soil-plant for different climates – an indication to reflect future development

Radionuclide	Modification factor (Temperate well =1)			
	Well		Rising ground water	
	Steppe(BS), Mediterranean (Csa) Boreal (Dfa)	Boreal (Dfc), Tundra (ET)	Temperate (Cfb), Boreal (Dfa)	Boreal (Dfc), Tundra (ET)
Cl-36	1	1	1	1
Ni-59	1	1	1	1
Se-79	10	1	1	0.3
Zr-93	0.5	1	1	1
Nb-94	0.5	1	1	1
Tc-99	10	0.1	1	0.1
Pd-107	0.5	1	1	1
Sn-126	0.5	1	1	1
I-129	0.5	1	10	10
Cs-135	0.5	50	0.5	50
Ra-226	0.5	0.5	1	0.5
Th-230	0.5	1	1	1
Pa-231	0.5	1	1	1
U-238	3	1	1	1
Np-237	5	1	1	1
Am-243	0.5	1	1	1
Pu-239	0.5	1	1	1

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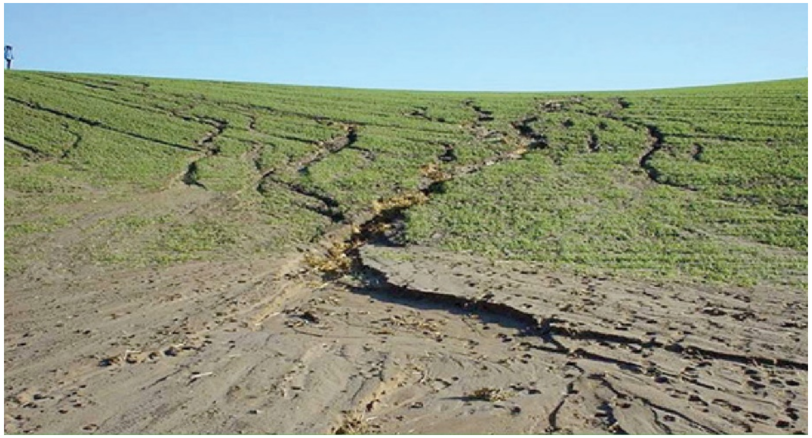
Erosion



A long-term process causing degradation and formation of soils

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Erosion:

Degradation of soil due to removal of soil material by wind and water

- **Erosion by water**
  - Kinetic energy of rain destroys soil aggregates
  - Soil will be transported downhill

- **Factors increasing water erosion**
  - Precipitation and contribution of heavy rain showers
  - Slope
  - High fraction of sand and silt
  - Low content of clay and organic matter
  - Poor vegetation

- **Relevance**
  - Up to 200 t/(ha*a) (=> 1 cm soil)

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Erosion by water

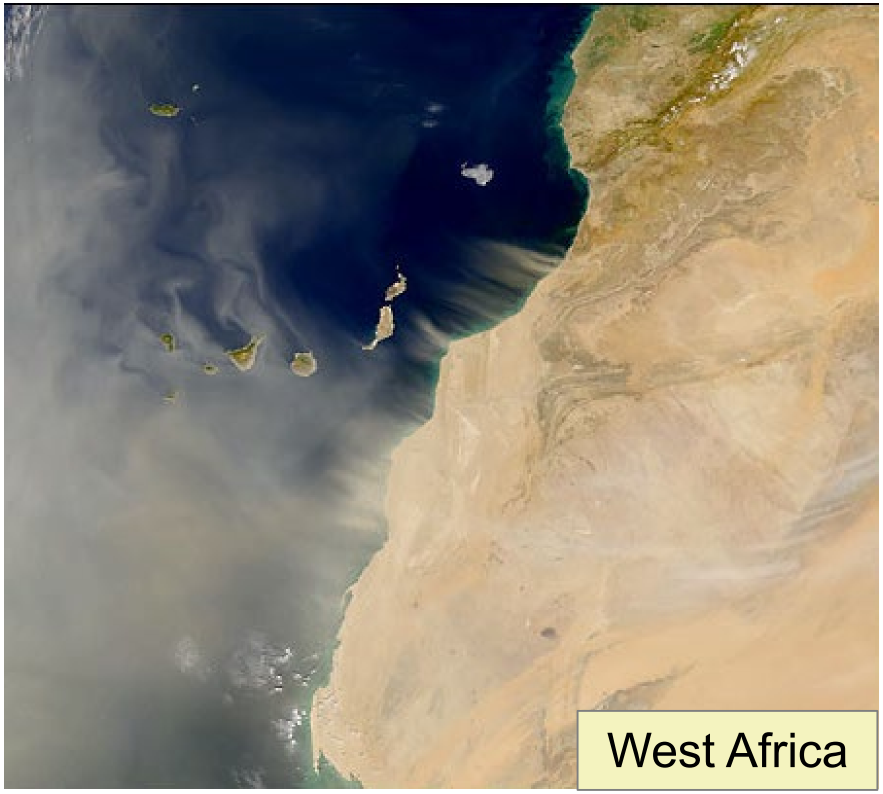
- Rain splash affects the soil surface
- Creeks in a sloping field

Soil erosion due to overgrazing

Yellow River – Soil material from China's loess areas

Wind erosion in Germany

- Wind erosion is non-continuous process
- The annual loss of soil varies widely
- The loss within 10 or 20 years may be caused by one event combining
  - Strong wind
  - Dry soil
  - No/little vegetation cover



Wind erosion during sandstorms

- Sand cloud in Texas
- An enormous sand cloud spreads from the Sahara over the East Atlantic

Loess in China formed by sedimentation of eroded material

Erosion is not a continuous process

- **Increasing relevance for high intensity events**
  - **Water erosion increases with**
    - … rainfall intensity and total amount of rainfall
    - … increasing slope
  - **Wind erosion increases with**
    - … increasing wind speed
    - … decreasing soil moisture

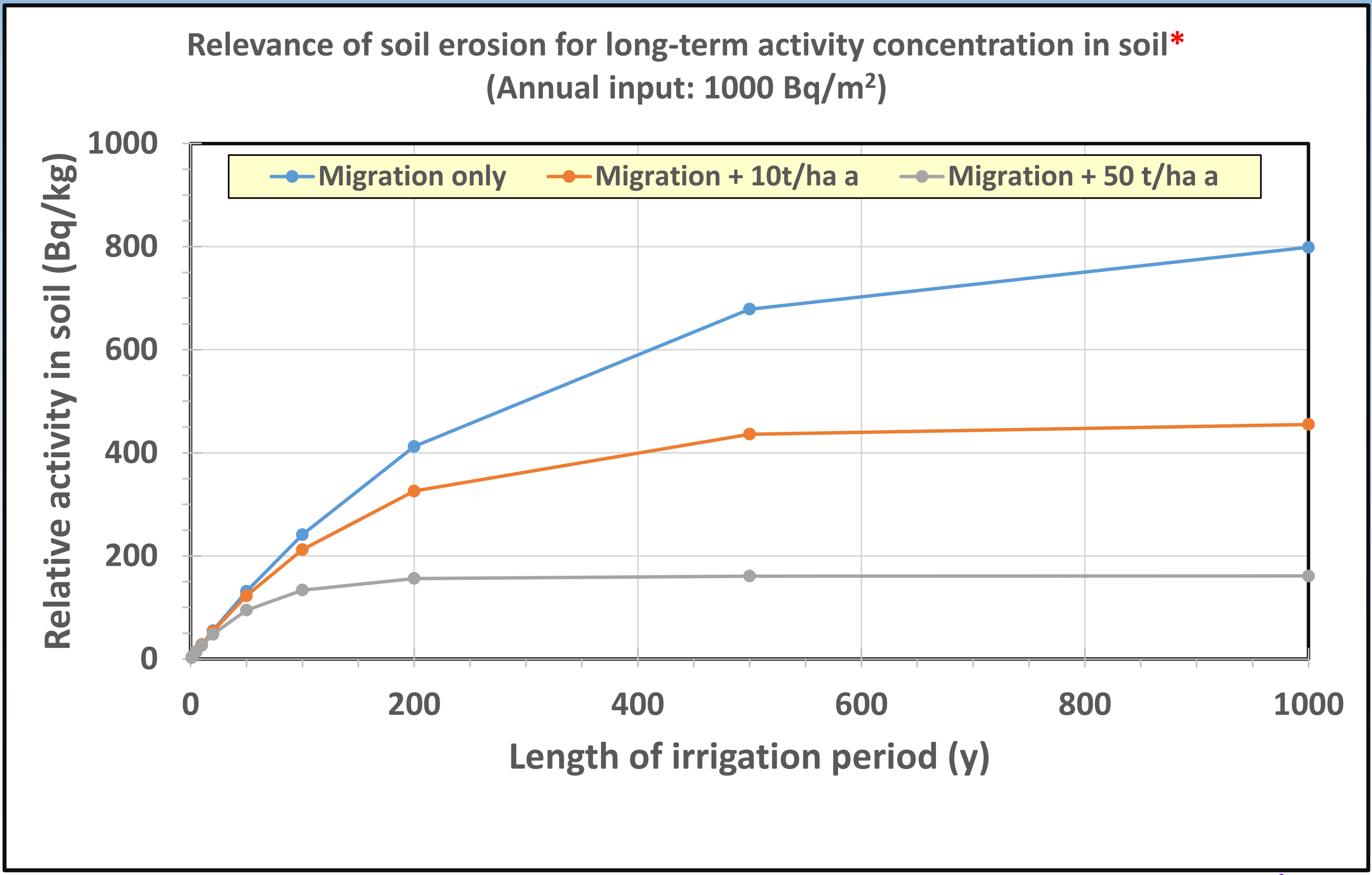
$\Rightarrow$ **Few events of**

- strong winds or

- heavy showers

cause most of the erosion

Erosion and migration



* Migration is is equivalent to a half-life in the upper 25 cm soil layer of 200 y

Implications of erosion for long-term safety assessments

- Water and wind **erosion will cause removal** of soil and of radionuclides bound to soil
- Erosion **will cause a wider distribution** of radionuclides that have been applied with contaminated irrigation water
- Erosion **will reduce the maximum** radionuclide concentrations of soil

Migration in soil (K_d-approach)

Transport of radionuclides in soil is driven by

- Water movement in the soil
- Sorption and desorption processes
- Derivation from the sorption coefficient K_d
- Retardation of the radionuclide transport compared to the water transport

$$\lambda_s = \frac{v_a}{L \cdot \left(1 + \frac{\rho}{\Theta} \cdot K_d\right)}$$

λ_s	=	Annual loss rate of the radionuclide from the layer L
v_a	=	Water velocity in the soil (m/a)
L	=	Thickness of the soil layer considered (m)
ρ	=	Soil density (kg/m^3)
Θ	=	Water content of the soil

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Migration of radionuclides attached to soil particles

K_d-approach

- Does not include the migration of radionuclides attached to soil particles

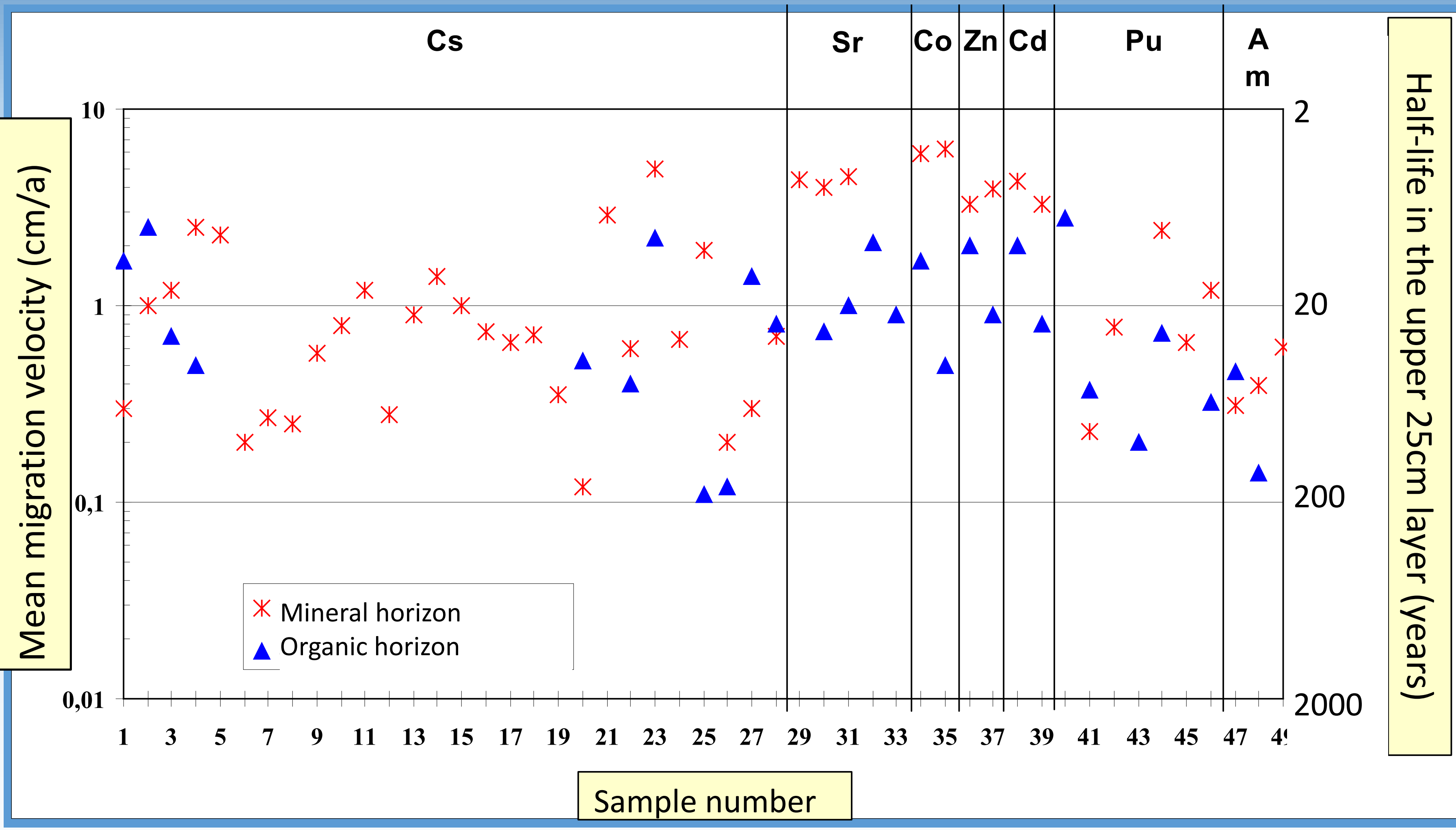
Relevant components

- Clay minerals, Fe-, Al- and Si-oxides
- Accumulation of clay in lower soil horizons can be observed in many soil types
- Migration is more important in acid soils (pH<6,5)
- Important in wet and periodically wet climates

Quantification of particle movement

- Of the order of 10 g/m² a
- It is a long-term process

Measured migration rates in soil (Bunzl, 92, 93, 94, 94a, 95)



Variation of K_d-values is larger than the measures migration rates

Modifications for different climates: Loss of soil due to erosion and migration to deeper soil layers

Climate	Half-life of radionuclides in soil due to erosion from the upper 25 cm (years)	Migration (relative values, temperate climate=1)
Temperate (Cfb)	1400	1
Mediterranean (Csa)	**350**	0.5
Steppe (BS)	**140**	0.2
Boreal (Dfa)	**350**	0.5
Boreal (Dfc)	1400	1
Tundra (ET)	1400	1

Erosion is particularly relevant for dry climates

Food intake rates for the different scenarios considered

Food item	Food intake rates (l a^{-1}, kg a^{-1})						
	Well. GW	Well, GW	Well	Well	Well, GW	Well	GW
	Temperate Cfb	Mediterra. Csa	Steppe BS	Boreal Dfa	Boreal Dfc	Tundra* ET	Tundra ET
Drinking water	730	**1100**	**1100**	730	730	730	730
Cereals	110	115	105	110	71	-*	-
Potatoes/roots	55	107	93	55	84	-	113
Leafy vegetables	13	56	34	13	51	-	7.7
Fruit vegetables	75	84	12	75	-	-	16
Milk	130	100	87	130	115	37*	37
Beef	30	28	22	30	72	110*	110
Pork	60	28	22	60	0	0	-
Lamb	0	**1,5**	**12**	0	0	0	-
Fish	1	0	0	1	14	0	**36.5**
Fungi	-	-	-	-	-	-	**7.7**
Berries	-	-	-	-	-	-	**16**
Reindeer	-	-	-	-	-	-	**110**
Reference site	Germany	Spain	Greece	Germany	Sweden	Kola peninsula	

* Water is only used as drinking water for humans and animals (no irrigation)

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Well scenario: Annual doses normalized to radionuclide concentration in well water

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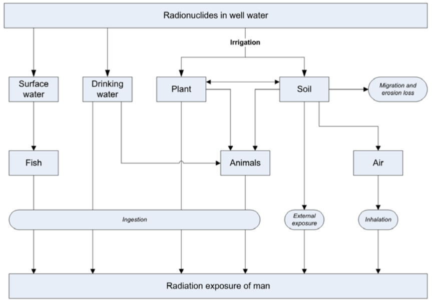

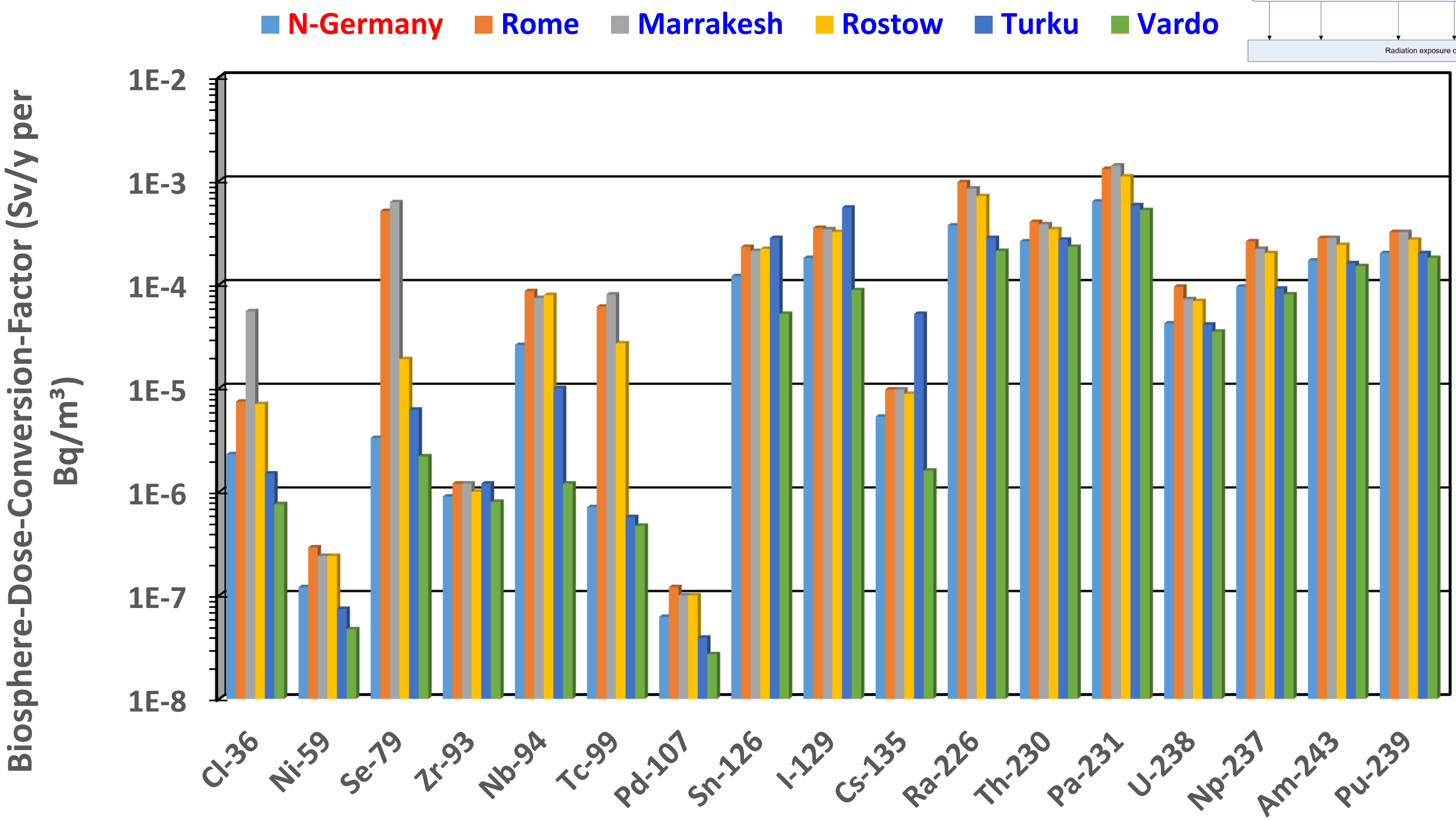

- Values increase with increasing amounts of irrigation water
- Little impact of climate for Sn-126, Ra-226, Th-230, Pa-231, U-238, Np-237, Am-243, Pu-239

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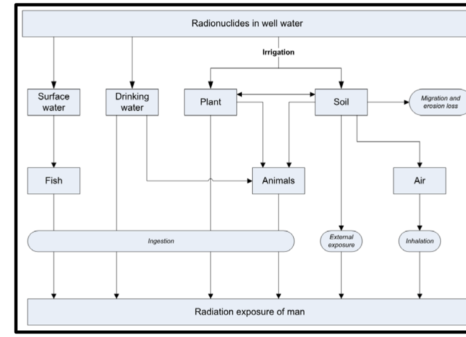
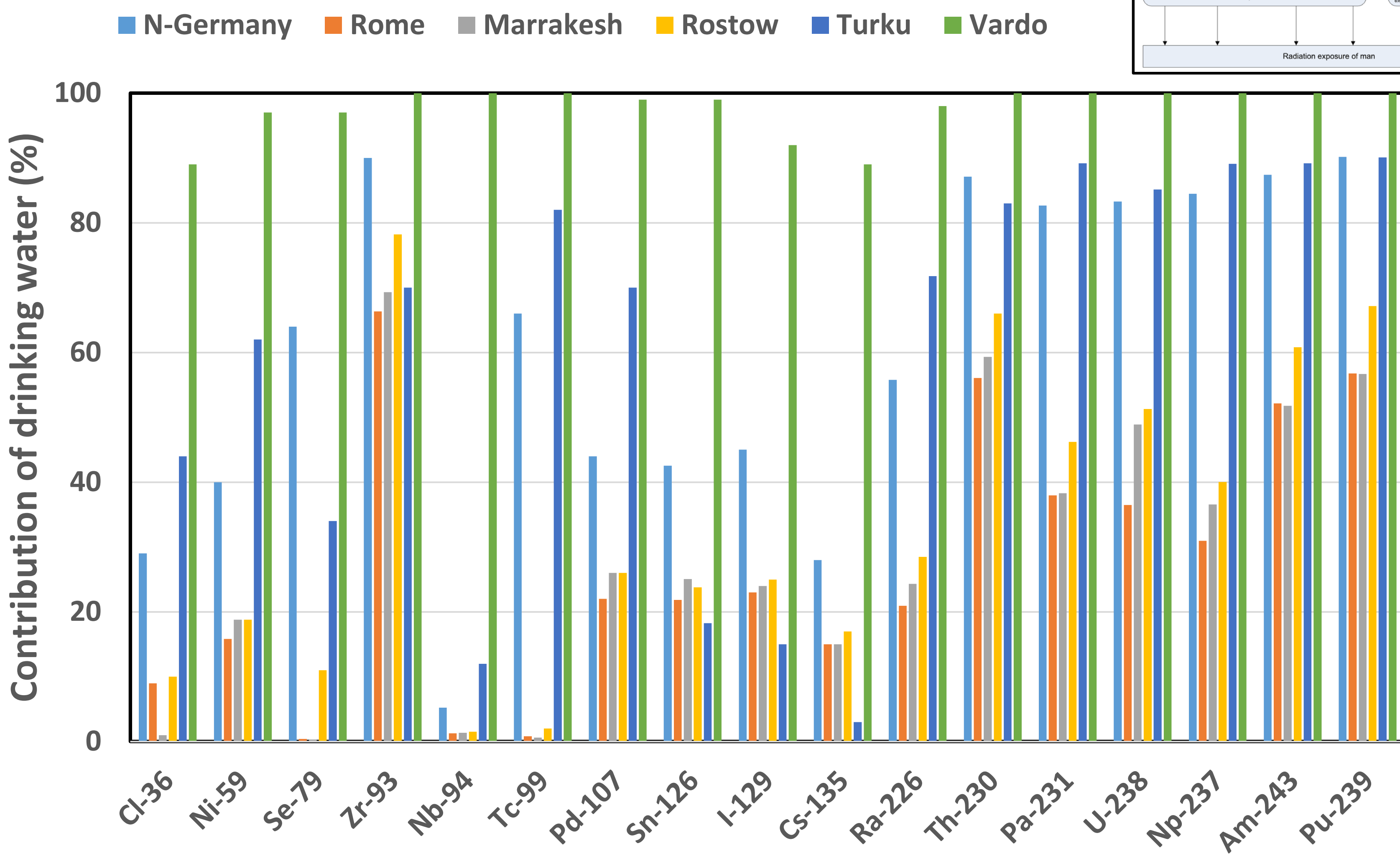
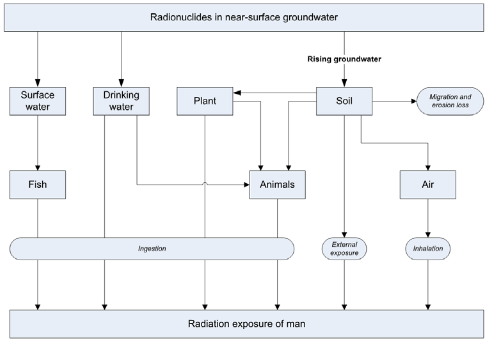
Well scenario: Contribution of drinking water to normalized exposure

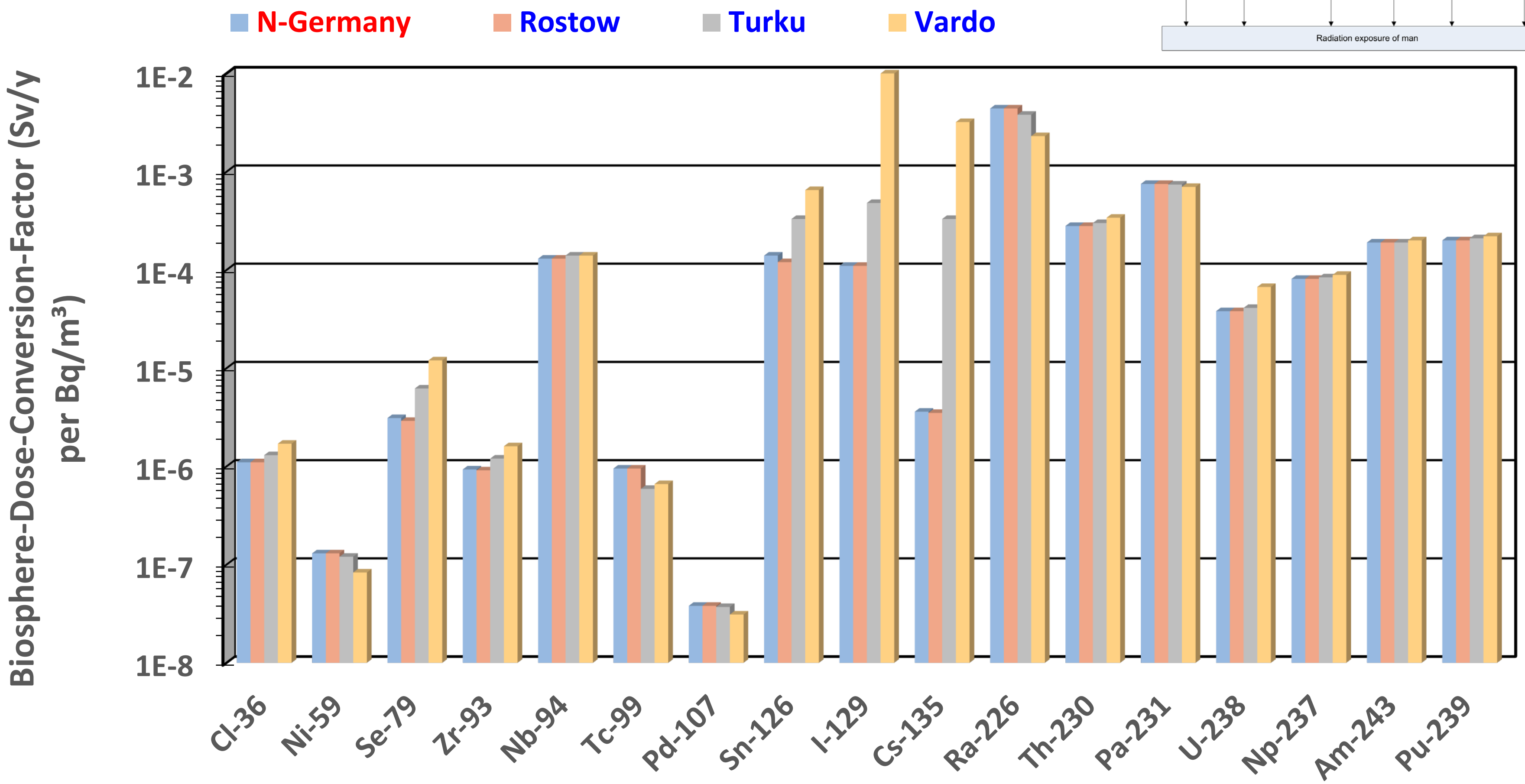




- Dominating contribution of drinking water for many radionuclides, in particular for colder climates

Scenario “Rising groundwater”: Annual doses normalized to radionuclide concentration in ground water





- Highest impact of climate for I-129 and Cs-137
- High availability of I and Cs in organic, acid soils

S U M M A R Y

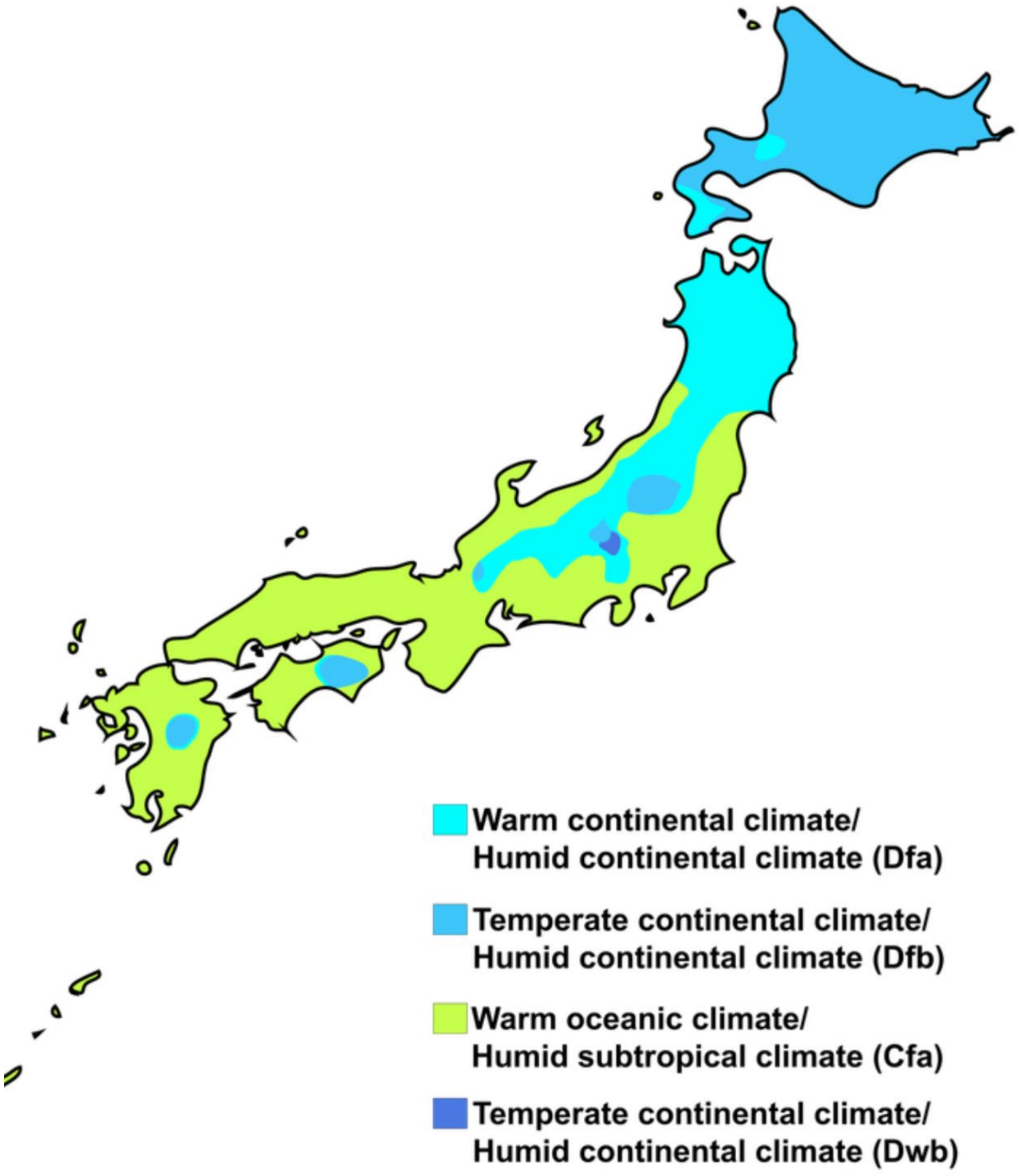
- **Environmental conditions will have changed…**
  - ….when radionuclides from a waste repository may enter the biosphere
- **Extrapolation of current conditions to the far future is too simple**
  - Inherently uncertain and speculative
- **Analogue approach**
  - Selection a range of sites define an envelope for future exposure conditions
  - Representing different climates, agricultural conditions
- **Long-term processes may cause relevant spreading of radionuclides in the environment**
  - Migration of radionuclides in soil
  - Erosion with wind and water
- **Results: Dose per unit radionuclide in water**
  - Potential exposures are higher for hot and dry climates
  - For redox-sensitive radionuclides (Se-79, Tc-99, I-129, Np-237) pronounced differences between climates
  - Ingestion is by far the dominating pathway
  - For many radionuclides the intake of drinking water dominates => little difference between climates

Additional information

Interaction of climate and land use

Climate	Region (examples)	Natural vegetation	Land use
Polar/subpolar	Polar region, High North of Eurasia and North America	Polar desert, tundra	No land use, Nomadism
Boreal	Northern Eurasia Northern North America	Lichens, coniferous forests	Nomadism, grazing, farming
Temperate, humid	Middle Europe, Eastern USA, Chile, **Japan**	Deciduous forests	Grazing, farming
Temperate, dry	Inner Asia, Central USA	Steppe	Nomadism, extensive grazing, Dry farming
Subtropical, rain during winter	Mediterranean region, California, SW Australia	Subtropical rain forest	Wine, olives, citrus, vegetables, wheat
Subtropical, rain during the whole year	Southern USA, SE Asia, East Australia, **Japan**	Steppe, savannah	Intensive farming
Dry Subtropical and Tropical Areas	Sahara, Arabia, North Chile, Inner Australia	Savannah	Nomadism, extensive grazing
Tropics, rain during the whole year	Northern Brazil, East India, Africa at the equator	Tropical rain forest	Farming

Climate of Japan



Times and development of soils

Object	Time
Half-life of ^{226}Ra	1 600 a
Half-life of ^{238}U	$4.5*10^9$ a
Time since the last ice-age	15000 a
Age of "young soils" in Middle and Northern Europe, North America (podzol, brown earth, para-brown earth)	1000 - 20000 a
„Old soils" in tropical region (Oxisols)	10^7 - 10^9 a
Age of humins	Up to 5000 a
Development of an A-horizon	10-1000 a

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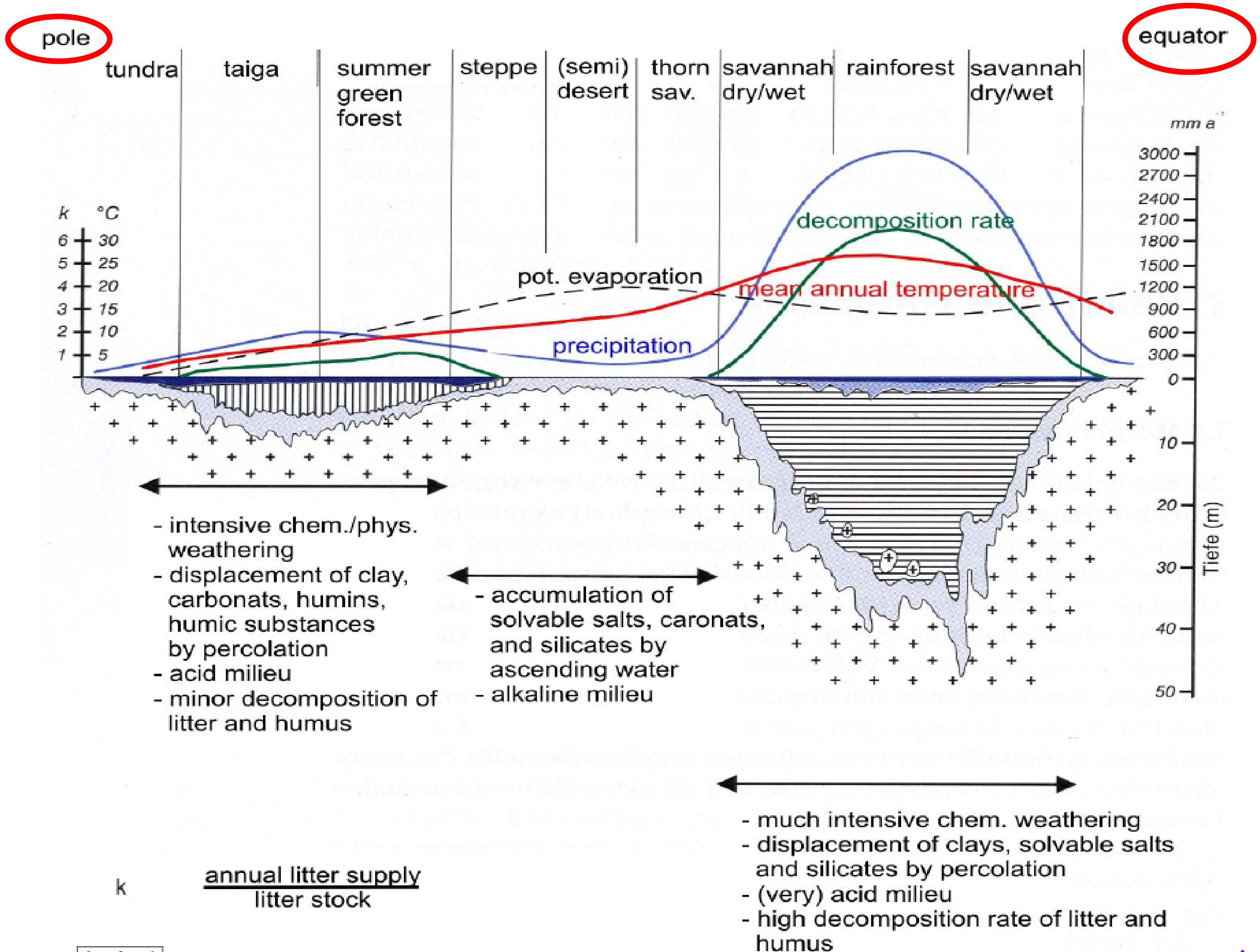
Interaction of soil and climate



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Soil processes and radionuclides in soil

Process	Tc	Se	I	Cl	Np	U
Leaching of Ca	-	-	-	-	Formation of mobile carbonate complexes	
Acidification (input of H^+, loss of cations)	Immobilisation		-	-	Mobilisation	Immobilisation
Leaching of clay	Migration of radionuclides attached to clay, relevant for strongly sorbed cations					
Podzolisation	Mobilization and precipitation of DOC, Al, and Fe					
Leaching of silicates	Increasing absorption of anions				Decreasing absorption of cations	
Erosion	Dislocation of radionuclides by water and wind					

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Examples for annual erosion (in t ha^{-1} a^{-1})

(A. Goudie 2007, Physical Geography)

	Natural land	Agricultural land	Uncovered soil
Australia	0-64	0.1-150	44-87
Belgium	0.1-0.5	3-30	7-82
China	0.1-2	150-200	280-360
India	0.5-5	0.3-40	10-185
UK	0.1-0.5	0.1-20	10-200
USA	0.03-3	5-170	4-9

10 t ha^{-1} a^{-1}: ca. 0.7 mm a^{-1}

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Bioturbation

- **Activity of soil organisms**
  - Borrowing animals as voles, rabbits, earthworm continuously mix the soil
  - Development of the root system has a mixing effect
- **Effects**
  - Improved aeration of soil
  - Formation of organo-mineral complexes
  - Increasing the soil stability and improving the porosity
  - Increase of porosity, formation of pathways allowing the transport of particle-bound radionuclides

Turnover of soil due to activities of soil animals

Animal	Substrat	Annual turnover (kg/m² a)
Earthworm	Garden soil	1
	Pasture	2-4
	Orchard/Forest	2-3
	Forest meadow	7-8
Ant	Forest	5
Isopods	Semi desert	0.15
Vole	Forest	1.2-12
Termites	Savannah	6
Ground squirrel	Semi dessert	0,15

Scheffer and Schachtschabel: Soil Science, 1982 (In German)

Turnover: a few percent per year

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General trends of effects of climate change on soil properties

Feature	Temperature increase		Temperatur decrease	
	Humid	Arid	Humid	Arid
Organic matter	↓	↑	↑	↑
Effect on radionuclide behaviour	Mobilisation	I m m o b i l i s a t i o n		
Weathering	↑	↓	↑	↓
Effect on radionuclide behaviour	Lower sorption of cations	No general trend	Lower sorption of cations	No general trend
pH-Wert	↓	↑	↓	-
Effect on radionuclide behaviour	Mobilisation	No general trend	Mobilisation	No general trend
Percolating water	↑	↓	↑	↓
Effect on radionuclide behaviour	Migration in soil increases	Migration in soil decreases	Migration in soil increases	Migration in soil decreases
Erosion	↑ Water erosion increases	↑ Wind erosion increases	↑ Water erosion increases	↑ Wind erosion increases