Assessing the impact of releases of radionuclides into sewage systems in urban environment – simulation, modelling and experimental studies

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LUCIA – NKS project participants

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LUCIA – the aim

- to assess the impact of liquid releases of radionuclides into the sewage systems in urban areas;
- to refine the modelling prediction tool(s);
- to test the prediction tools for impact assessment;
- advice and recommendations on risk assessments following radionuclide releases (urban);

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LUCIA - performance

- Predictions based on the model LUCIA
- Focus on the releases from hospitals
- Experimental studies conducted
- Dosimetric studies

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Why focussing on the sewer system?

- Sewage sludge is a sensitive indicator of contaminants entering the environment (*Erlandsson et al 1978 in Water Air and Soil Pollut.9.,);*
- In urban areas wastewater transported to the sewage plant;
- Sewage system acts like a ”kidney” in urban areas;
- Patients treated in hospitals with radiopharmaceuticals;
- Contaminated excreta fed directly into the sewage systems;

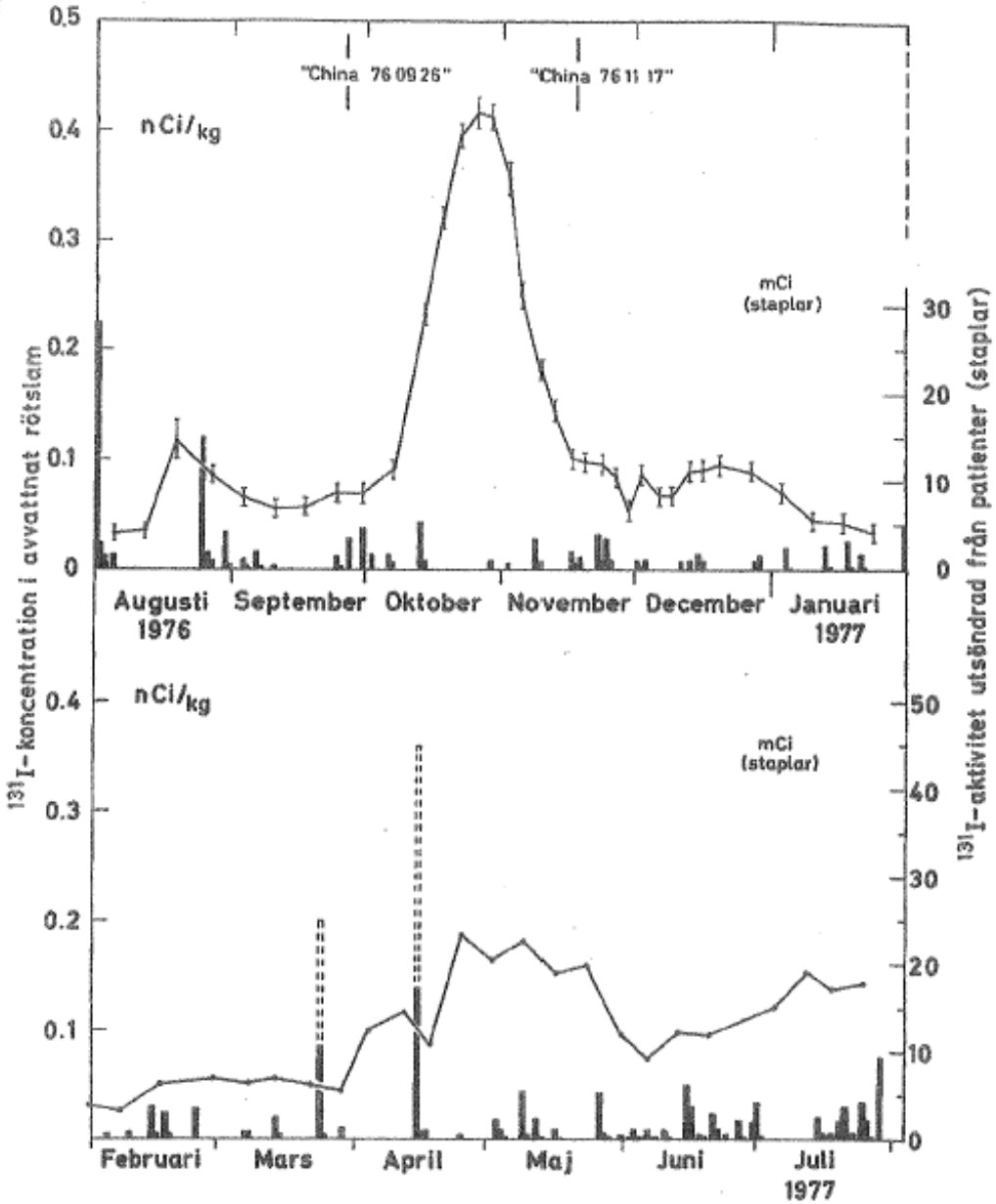

Fig. 4. Jod-131 koncentrationen i det avvattnade rötslammet under ett år. Spalarna representerar utsläpp av jod-131 från patienter som fått ^{131}I-terapi på Allmänna sjukhuset. Två kärnvapenprov är också markerade.

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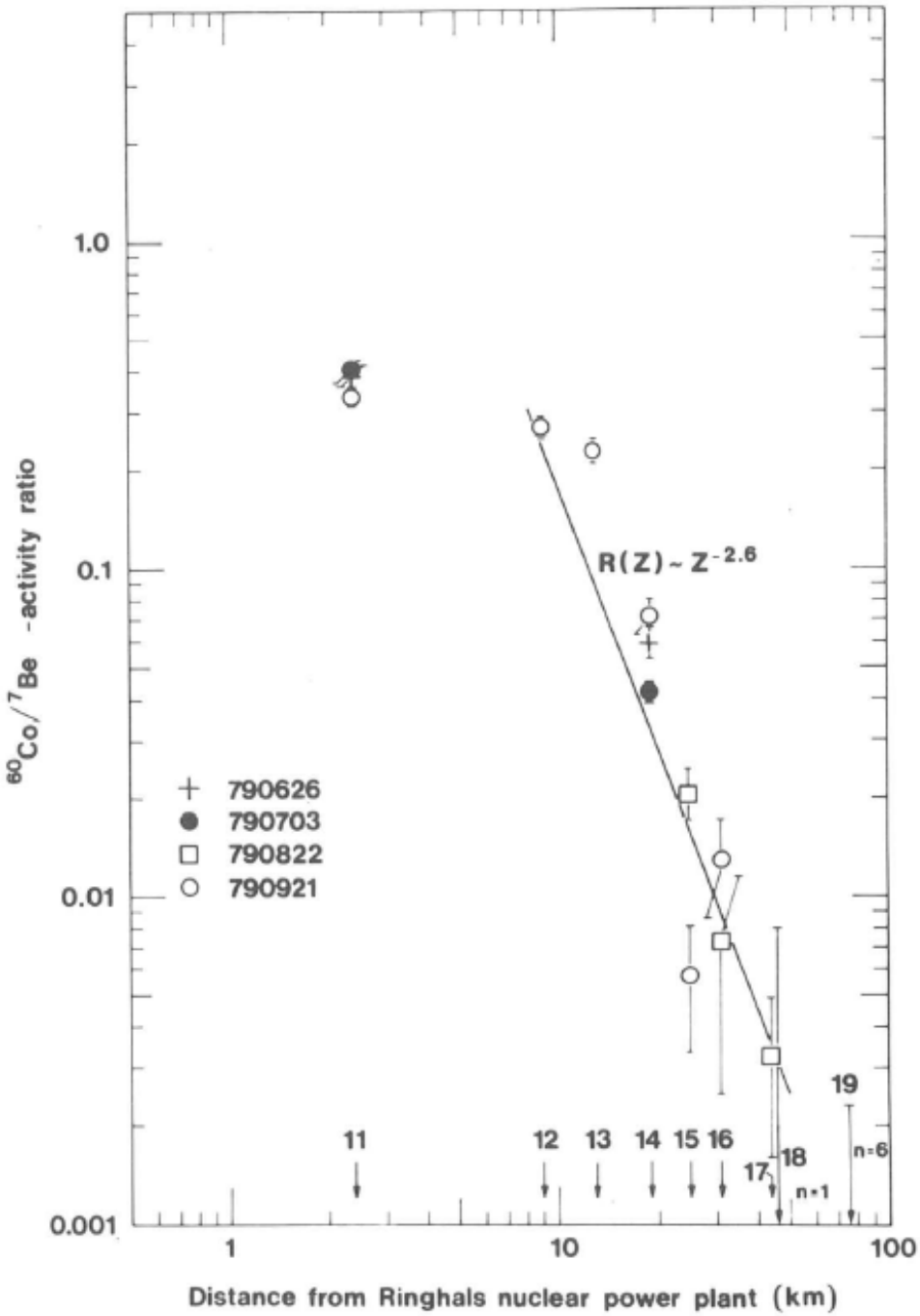
^{60}Co from the nuclear power station at Ringhals



Ingemansson, Mattsson and Erlandsson, 1981

LUCIA – presumptions

- The design of sewage plants is relatively similar between the Nordic countries;

- The process of wastewater treatment is relatively similar between the Nordic countries;

- The treatments at hospitals are relatively similar between the Nordic countries;

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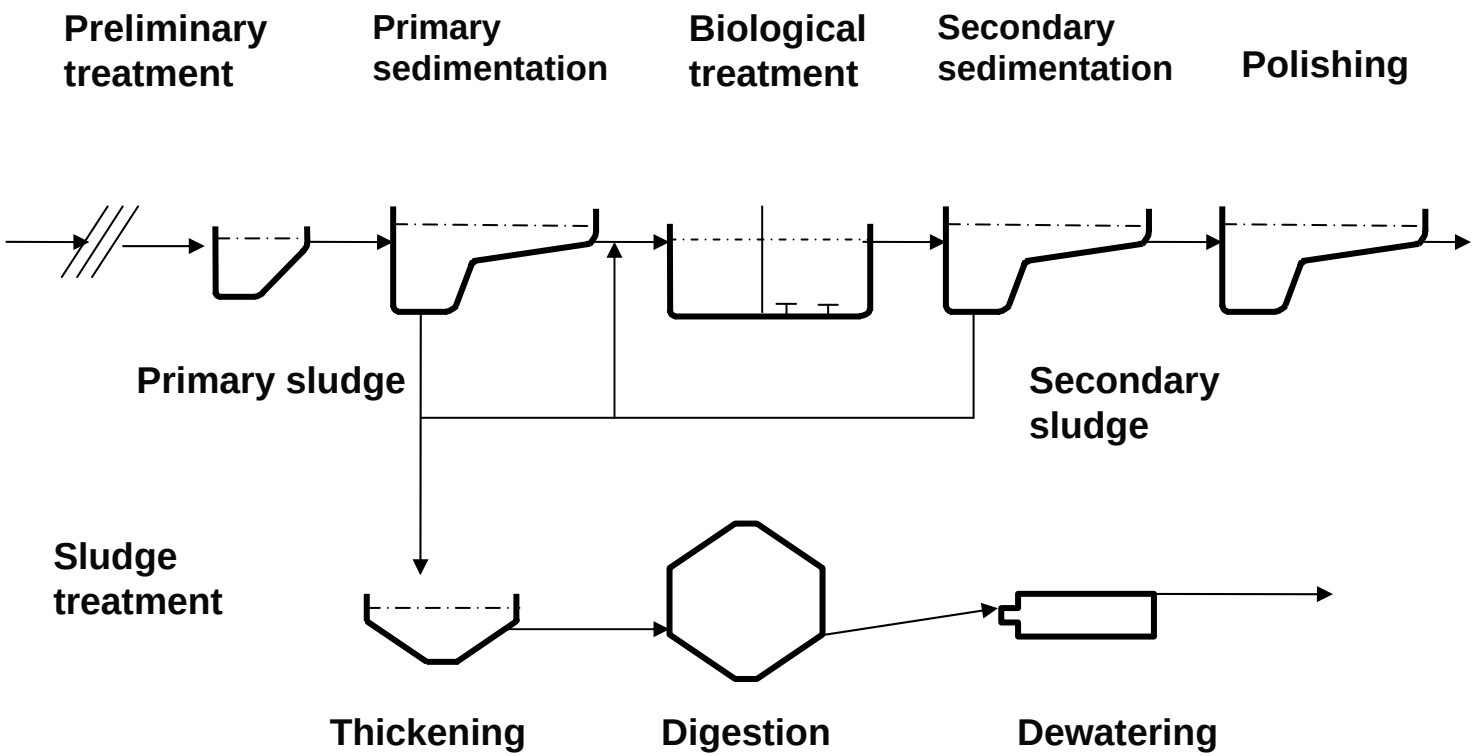
Waste-water treatment plant



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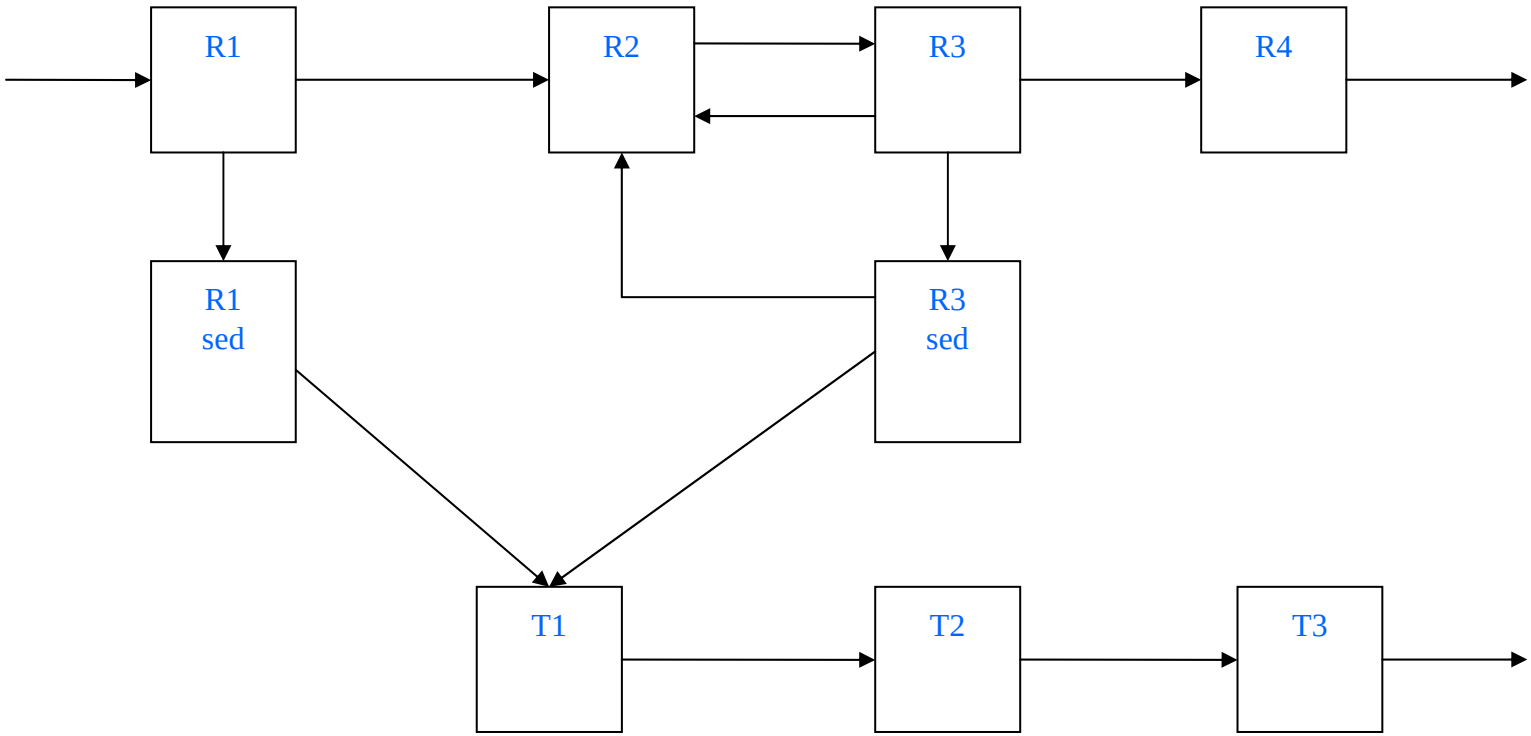
Conceptual LUCIA model



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Compartments

Compartment	Plant components	State variable
R1	Primary sedimentation (water)	Nuclide inventory (tot) dissolved
R1 sed	Primary sedimentation (precipitated sludge)	Nuclide inventory (tot) in prec. sludge
R2	Basins (biological)	Nuclide inventory (tot) dissolved
R3	Secondary sedimentation (water)	Nuclide inventory (tot) dissolved
R3sed	Secondary sedimentation (precipitated sludge)	Nuclide inventory (tot) in prec. sludge
R4	Basin (final, water)	Nuclide inventory (tot) dissolved
T1	Thickener	Nuclide inventory (tot) in the component
T2	Digester	Nuclide inventory (tot)
T3	Centrifuge and sludge storage	Nuclide inventory (tot)

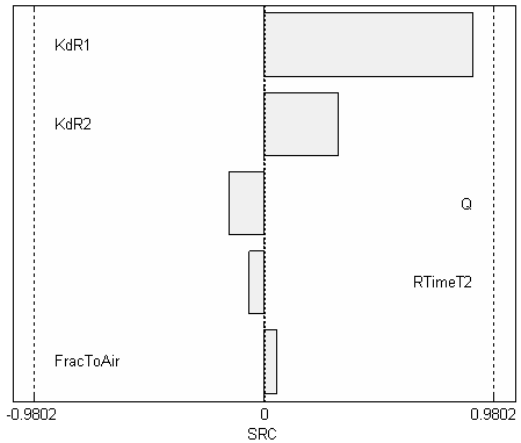
Model assumptions

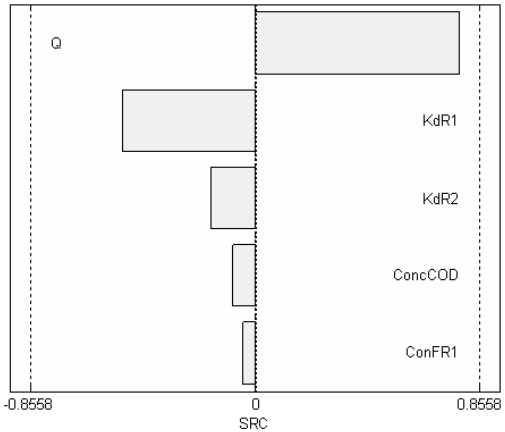
- Compartment model -> instantaneously homogeneous mixing in compartments of matter entering
- Transfer from donor to acceptor compartment proportional to inventory in donor compartment
- The model assumes steady state conditions for the wastewater, solids and organic material in the system.
- Kd values for organic soil were used because of lack of literature values
- Distributions were assigned to model parameters

Sensitivity analysis

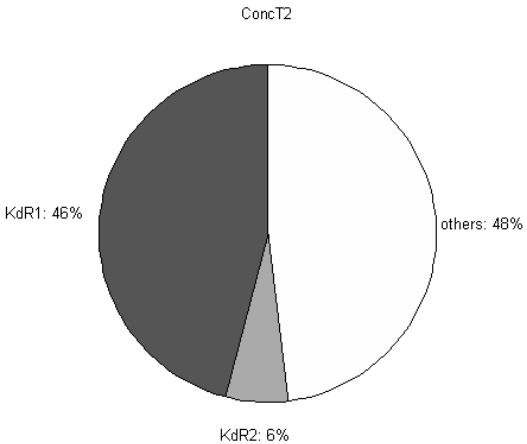
- Carried out using the tool EIKOS (Ekström *et al.* 2006)
- Done for I-131 with endpoints being the *concentration in digested sludge* (T2) and the *efficiency* of the waste-water treatment (defined as: activity conc. in incoming water/activity conc. in water released from plant)
- Contribution of model parameters to variance of predictions was estimated using Total Sensitivity Indexes calculated with the Extended Fourier Amplitude Sensitivity Test, EFAST

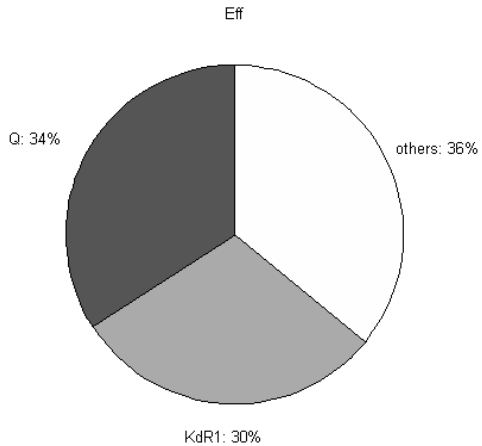
Kd-sensitivity





Spearman Rank correlation coefficient





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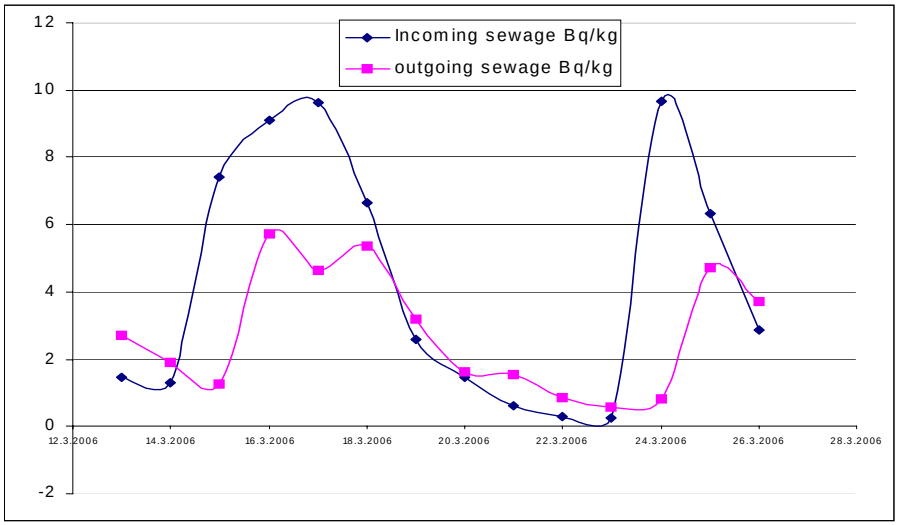
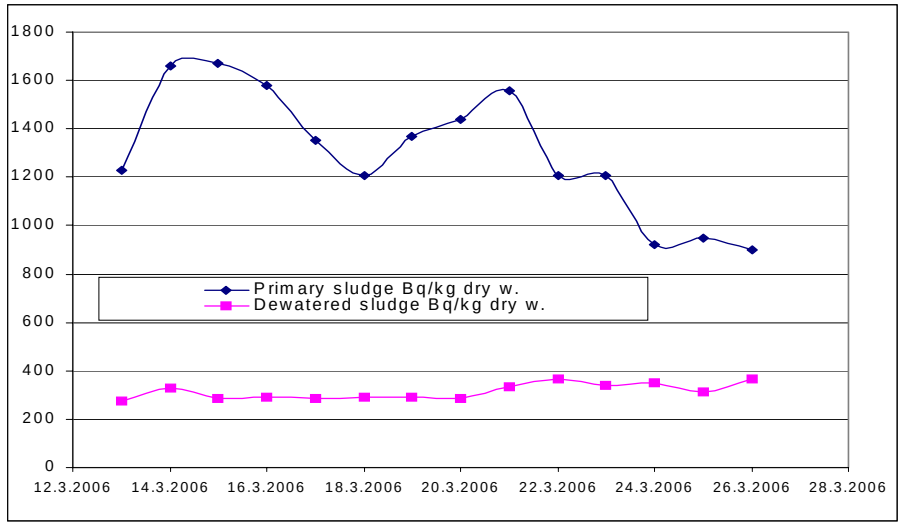
^{131}I in sewage and sludge at Viikinmäki, in Helsinki





Administered activity to patients

Date	Nuclide	MBq
7.3.2006	^{131}I	3700
10.3.2006	^{131}I	7400
15.3.2006	^{131}I	3700
17.3.2006	^{131}I	3700
24.3.2006	^{131}I	3700

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Dose assessments

- Releases of Tc-99m and I-131
- Hypothetical individuals
- Sewage worker
- Member of the public exposed to water
- Member of the public exposed to contaminated sludge
- LUCIA model
- Parameter values from questionnaire and IAEA 2001.

Questionnaire (parameter values)

- Geographic location
- Hospitals served
- Recipients
- Water and sludge treatments
- Number of equivalent persons/year
- Water flux (m^3/day)
- Sludge (Kg dw/day)
- Process parameters (COD, solids, volumes…)
- Residence times

Annual release rates to the plants

Country		Sweden	Finland	Norway	Denmark	Iceland
Sewage plant		Kungsängverket	Viikinmäki	VEAS Vestfjorden avlöpsselskap	Renseanlaeg Vest	Skólpa Klettagardar
Release rate, Bq/y	I-131	1.3E+11[1]	7.3E+11[2]	9.0E+11[3]	2.2E+11[4]	1.9E+11[5]
	Tc-99m	1.4E+12[1]	2.4E+12[2]	7.5E+12[3]	-	-

Concentrations in water and sludge (LUCIA)

Sewage plant	Nuclide	C_{water}, Bq/m^3	C_{sludge}, Bq/kg dw
Kungsängsverket	Tc-99m	5E+03	2,9E-01
	I-131	6,6E+03	9,4E+01
Viikinmäki	Tc-99m	4,2E+03	3,7E-01
	I-131	7E+03	9,4E+01
VEAS Vestfjorden	Tc-99m	5,9E+04	2,4E-01
	I-131	8,1E+03	7,2E+01
Renseanlaeg Vest	I-131	7,3E+03	8,8E+01
Skólpa Klettagardar	I-131	6,1E+03	1,7E+02

Annual dose rates (Sv/a) from water and fish

Sewage plant	Nuclide	Dose $_{water}$	Dose $_{fish}$	Dose $_{total}$
Kungsängsverket	Tc-99m	6,6E-08	6,6E-08	1,3E-07
	I-131	8,7E-05	1,8E-04	2,6E-04
Viikinmäki	Tc-99m	5,6E-08	5,6E-08	1,1E-07
	I-131	9,3E-05	1,9E-04	2,8E-04
VEAS Vestfjorden	Tc-99m	7,8E-07	7,8E-07	1,6E-06
	I-131	1,1E-04	2,1E-04	3,2E-04
Renseanlaeg Vest	I-131	9,7E-05	1,9E-04	2,9E-04
Skólpa Klettagardar	I-131	8E-05	1,6E-04	2,4E-04

Annual doserates (Sv/a) for sewage workers

Sewage plant	Nuclide	Dose $_{ext}$	Dose $_{inh}$	Dose $_{total}$
Kungsängsverket	Tc-99m	4,3E-09	1,7E-16	4,3E-09
	I-131	3,8E-06	3,3E-11	3,8E-06
Viikinmäki	Tc-99m	5,6E-09	2,2E-16	5,6E-09
	I-131	3,8E-06	3,3E-11	3,8E-06
VEAS Vestfjorden	Tc-99m	3,6E-09	1,4E-16	3,6E-09
	I-131	2,9E-06	2,6E-11	2,9E-06
Renseanlaeg Vest	I-131	4,2E-06	3,6E-11	4,2E-06
Skólpa Klettagardar	I-131	7E-06	6E-11	7E-06

Annual dose rates (Sv/a) related to farming

Sewage plant	Nuclide	$Dose_{total}$, Sv/a
Kungsängverket, Sweden	Tc-99m	1,3E-13
	I-131	2,0E-08
Viikinmäki, Finland	Tc-99m	1,7E-13
	I-131	3,0E-08
VEAS Vestfjorden avlöpsselskap, Norway	Tc-99m	1,1E-13
	I-131	2,3E-08
Renseanlaeg Vest, Denmark	I-131	2,9E-08
Skólpa Klettagardar, Iceland	I-131	6,4E-9

NKS - questions to be answered

- What work has been carried out?
- Why has the work been carried out?
- Who will use the results from the work that has been carried out?

Conclusion (model)

- Screening methodology not applicable (IAEA 2001)
- ”Realistic” assessments needed
- Requires many data, not always available
- A process orientated model was proposed
- Combine the Activated Sludge Model No.1 with LUCIA (K_d)
- Able to estimate concentrations and retention time of the sludge

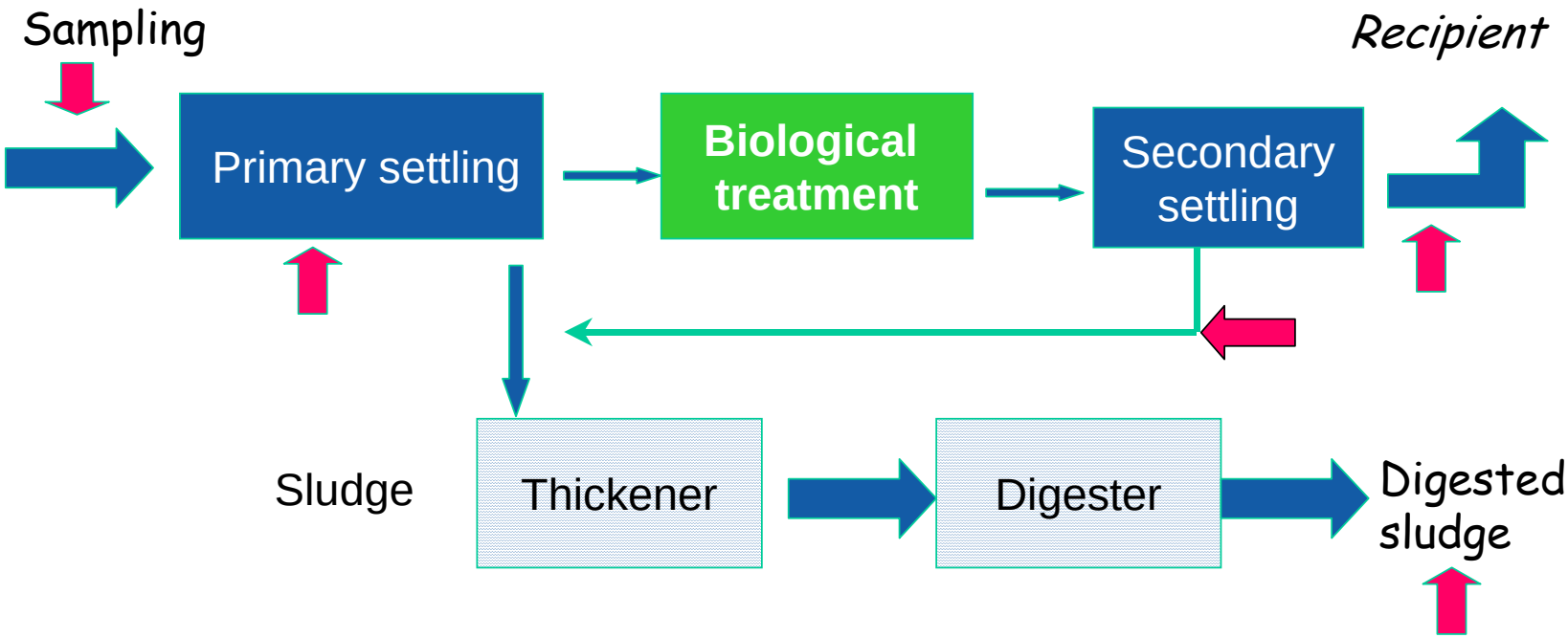
Conclusion (dose assessments)

- Similarities between the Nordic countries;
- Annual doses to the public insignificant (< 10 μSv/a);
- Dilution important for I-131 releases;
- Doses associated with farming lower compared to water pathways;
- Doses from Tc-99m to sewage workers were insignificant;
- Doses from I-131 (external) approach 10 μSv/a if uncertainties are considered;

Conclusions and current status

- Many regulated discharges to sewer, and there are no limits or the limits can be relatively high (> 1 TBq for radioiodine);
- Regulators have used very simple dose assessments tools in the past (MS Excel);
- The dose implications are generally very low (short lived);
- Further work to assess the partitioning of radionuclides between sludge and effluent and to improve understanding of exposure of sewage workers;
- Require to demonstrate BAT for uses;

Sampling and measurements



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Activities of ^{131}I in sewage and sludge compares to the administered activities

Date	Administered activity		Incoming sewage	Outgoing sewage	Primary sludge	Dewatered sludge
	MBq	Form	MBq d^{-1}	MBq d^{-1}	MBq d^{-1}	MBq d^{-1}
7.3.2006	3700	MIBG				
10.3.2006	7400	Iodide				
13.3.2006			287	533	73.8	16.6
14.3.2006			278	402	99.7	19.8
15.3.2006	3700	Iodide	1539	259	100.3	17.0
16.3.2006			1920	1210	94.9	17.6
17.3.2006	3700	Iodide	2097	1013	81.1	17.2
18.3.2006			1385	1118	72.3	17.3
19.3.2006			542	671	82.1	17.3
20.3.2006			305	341	86.2	17.0
21.3.2006			127	329	93.3	20.1
22.3.2006			61.6	180	72.4	22.1
23.3.2006			57.2	128	72.5	20.2
24.3.2006	3700	Iodide	2037	172	55.2	20.9
25.3.2006			1306	970	56.8	18.8
26.3.2006			597	769	54.1	21.9

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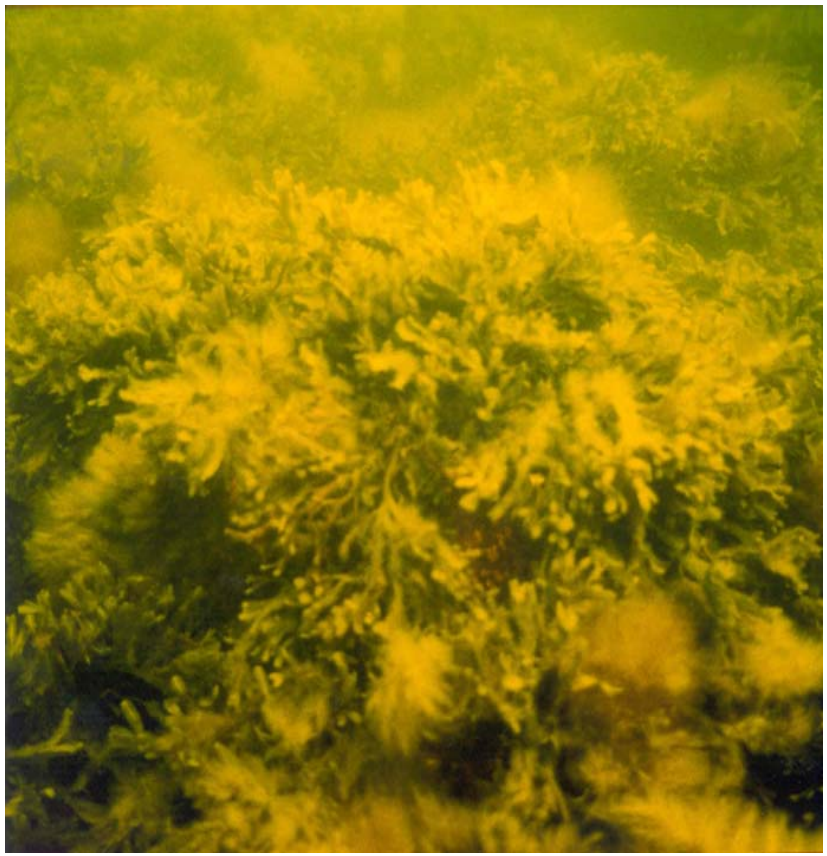
Brown algae, *Fucus vesiculosus*



Sampling date	^{131}I Bq kg^{-1} d.w.	Sampling Depth m
23.11.1983	1.0	
27.9.1985	0.4	
30.7.1992	33	
14.9.1995	210	
8.7.1999	186	
11.6.2007	102±12	2 -3.5
11.6.2007	52±7	3.5 -4.5

Thank you!

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Samples taken for LUCIA project

- In 2006 during two weeks in the Viikinmäki wastewater treatment plant
  - Incoming water
  - outgoing water
  - primary sludge
  - dried digested sludge
- In 2007
  - Two air samples in the wastewater treatment plant
  - Two Fucus vesiculosus samples in the sea area near Helsinki

Radiopharmaceutical administered to patients in Finland for therapeutic purposes (in 2006)

Radiopharmeceutical	Application	Administered activity, per patient	
		mean, MBq	range MBq
^{131}I-iodide	thyrotoxicosis	367	185-555
^{131}I-iodide	thyroid carcinoma	3816	2700-5500
^{32}P-phosphate	blood disease	144	111-185
^{90}Y-citrate	joint	220	
^{90}Y Zevalin	cancer metastases	1173	920-1596
^{153}Sm EDTMP	bone metastases	2956	2040-3000

Removal of nuclides from wastewater during the treatment process

Accumulation into sludge

- I-131 (iodide) 20 - 30 %
- P-32 > 90 %
- In-111 c. 50 %
- Tl-201 most part
- Sm-153 EDTMP most part ?
- Tc-99m ?

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Air samples

- The ventilation system of the treatment process area at the Viikinmäki wastewater treatment plant transports all emission to a tall chimney from where it is rapidly diluted into the atmosphere.
- Two air samples were taken from the discharge air in a pipe using activated carbon and glass fibre filters. The samples were collected from the air from the wastewater treatment process, excluding the air from the sludge process.

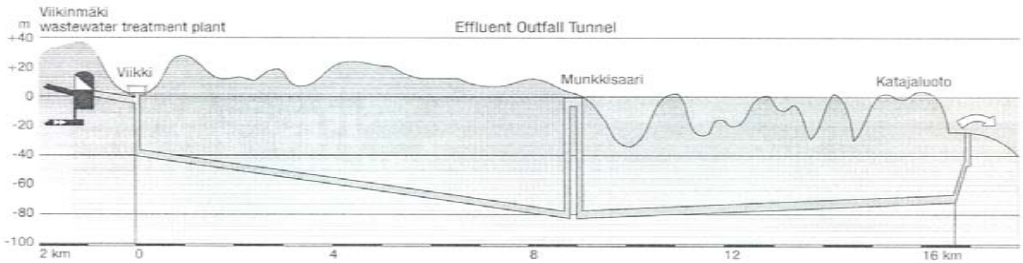
- The activated carbon and the filter were measured gammaspectrometrically in a plastic vial.
- A small amount of ^{131}I was detected in the activated carbon filters.

- In the glass fibre filters the activity concentration of ^{131}I was below the detection limit (MDA 53 - 116 µBq m^{-3}).

^{131}I in the air in the Viikinmäki wastewater treatment plant

Administered activity to a patient Date	MBq	Sampling period	Sampling speed l s^{-1}	the amount of air m^{-3}	^{131}I in outflow air µBq m^{-3}	kBq d^{-1}
19.3.2007	7.4					
20.3.2007	3900					
21.3.2007	7.4					
		22.3.07 14:30 - 23.3.07 12:10	3.9	302	191 ± 103	1.8
26.3.2007	3890					
	1137					
	8.04					
28.3.2007	3800	28.3.07 14:13 - 29.3.07 12:35	3.8	317	738 ± 162	7.1

Fucus vesiculosus samples



- The final effluent from the Viikinmäki plant flows through an outfall tunnel to a discharge area in the sea 8 km off Helsinki
- *Fucus vesiculosus* samples were collected in the sea, near the outfall tunnel
- The samples were measured directly gammaspectrometrically in Marinelli-beakers without any pretreatment.
- After the measurement the *Fucus* sample was dried at 105ºC and dry weight of the sample was determined.

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Concentrations of gamma emitting radionuclide in Fucus vesiculosus

Sampling date	Sampling depth	Bq kg^{-1} d.w.			
	m	^{7}Be	^{40}K	^{131}I	^{137}Cs
11.6.2007	2-3.5	29±5	901±108	102±12	19.6±2
11.6.2007	3.5-4.5	48±9	874±140	52±7	19.8±3

Study 2006 - 2007

- 6 samples daily for 13 days (13-29 Sep. 2006)

1. Water samples: incoming and outgoing
2. Sludge samples: primary and digested

- 24-hour composite samples, except for the weekends
- Totally 76 samples

- 9 TLD measurements (Sep. – Nov. 2007)

TLD dose measurements

9 TLDs were placed at the plant for 2 months:

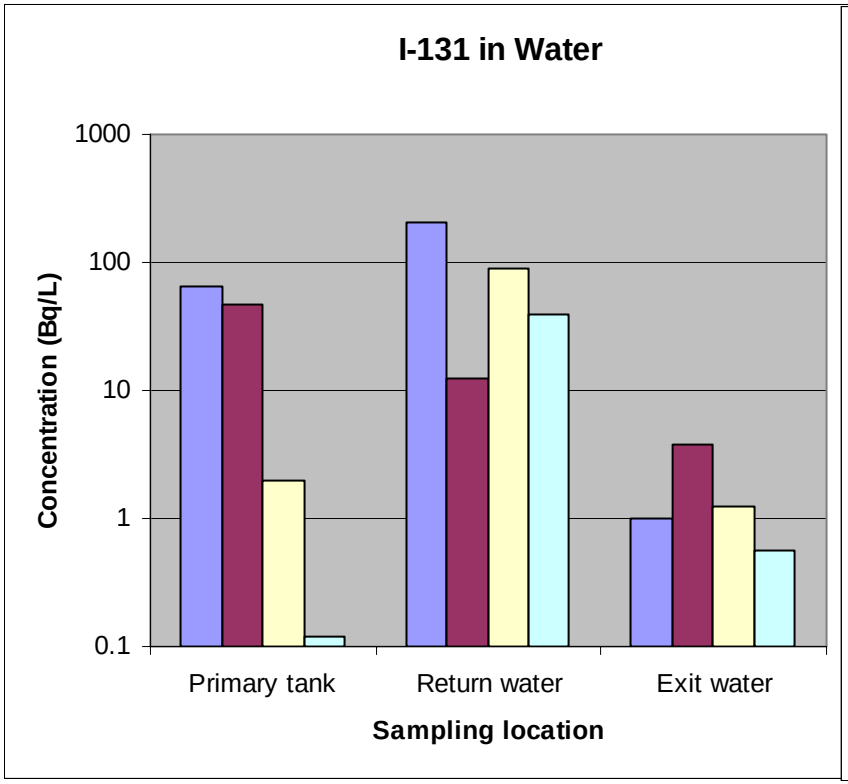
- 2 TLDs at screens
- 3 TLDs at dewatering filters
- 2 TLDs at drum thickeners
- 2 TLDs at process hall 8

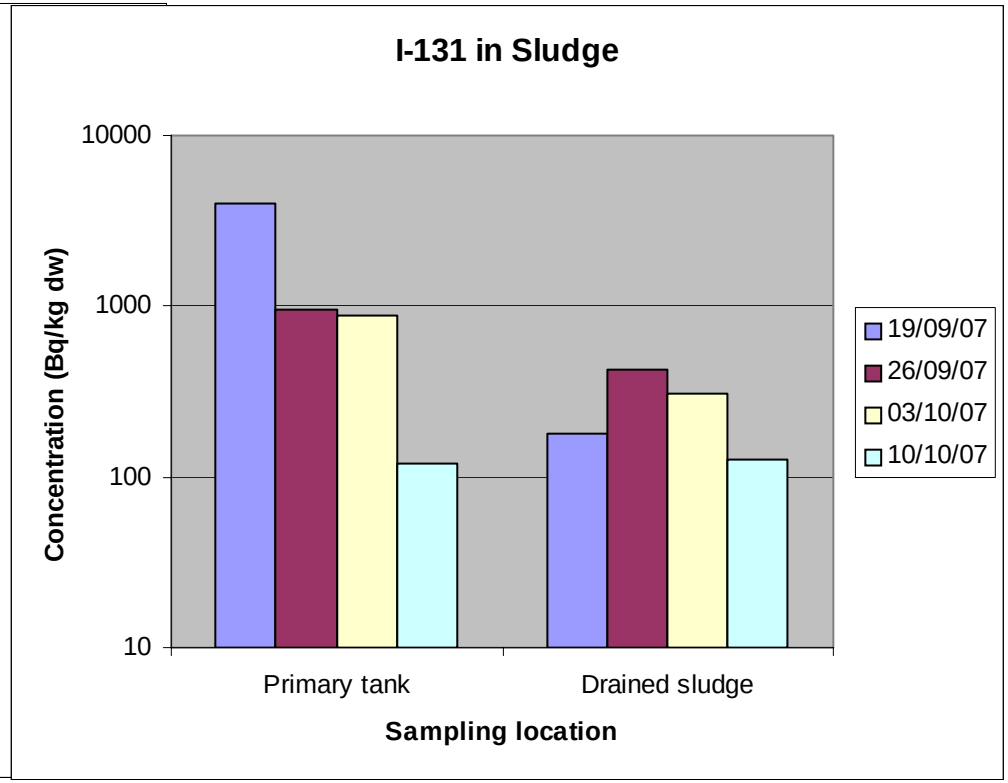
- Nothing was detected by TLDs

Schedule of radioiodine treatments at Aalborg Hospital

Date	Nuclear Medicine Dept. Diagnostic (MBq)	Oncology Dept. Ablation (MBq)
03-09-2007	1200	
04-09-2007		3700
10-09-2007	3600	
17-09-2007	1600	
18-09-2007		3700
21-09-2007		3700
24-09-2007	2200	
25-09-2007		3700
30-09-2007		3700
01-10-2007	1100	
08-10-2007	600	
15-10-2007	2200	
22-10-2007	600	
29-10-2007	3500	

Iodine-111 in sludge and water at Aalborg WWTP





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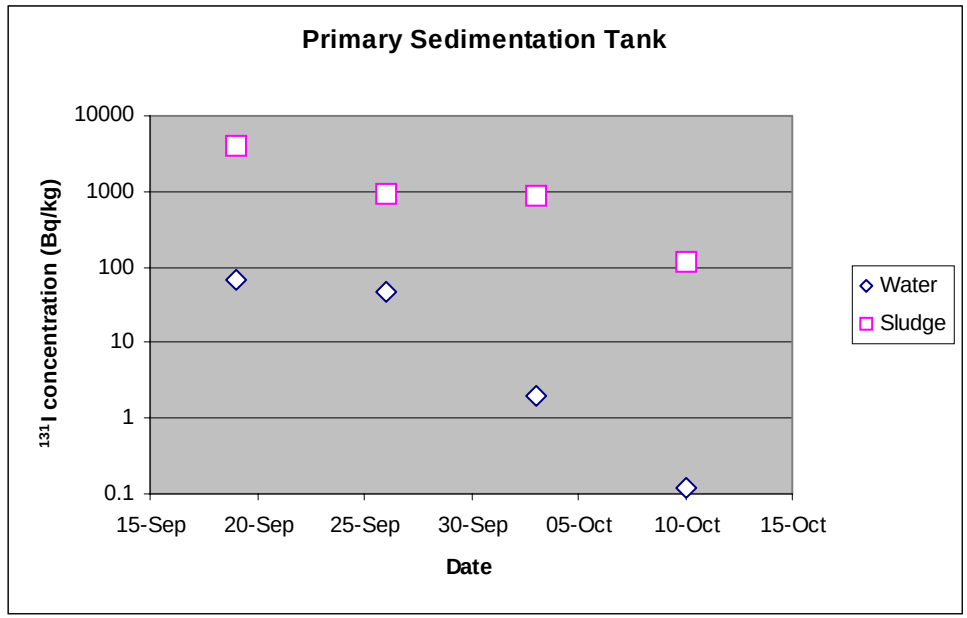
Distribution coefficient K_d for ^{131}I estimated from primary tank

- Observations of ^{131}I in water and sludge used to derive K_d-values though assumptions may not hold
- Derived K_d-values in the range 20-1000 L/kg, geometric mean value of 150 L/kg
- Reference K_d-values for marine environment of 200 L/kg for open ocean and 70 L/kg for ocean margin (IAEA TRS-422, 2004)



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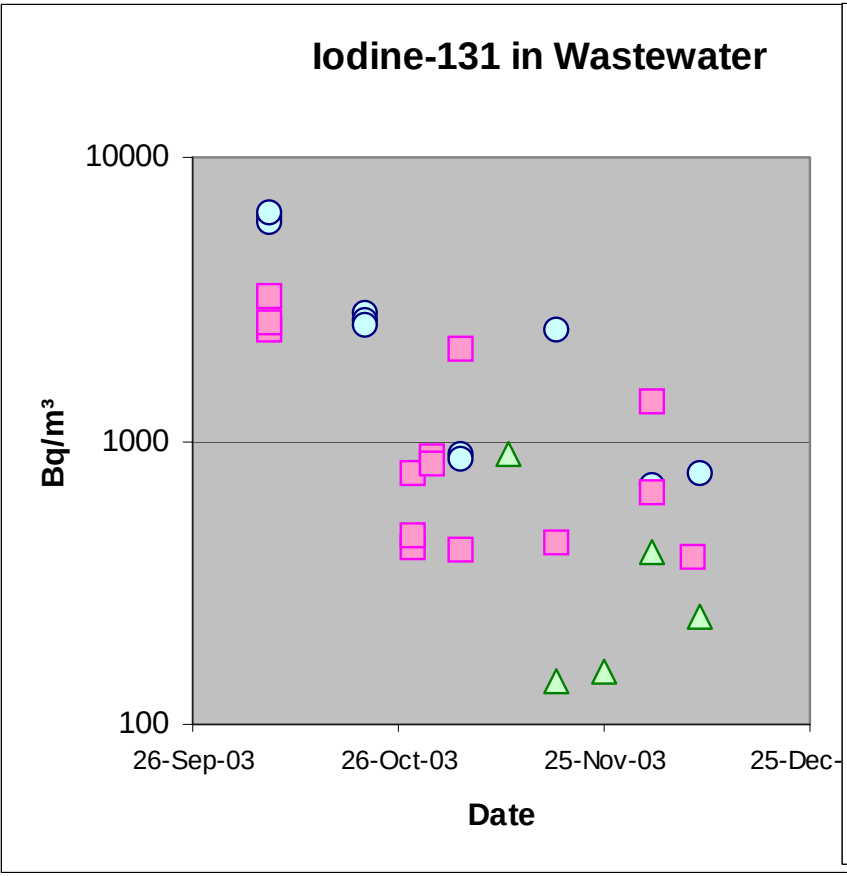
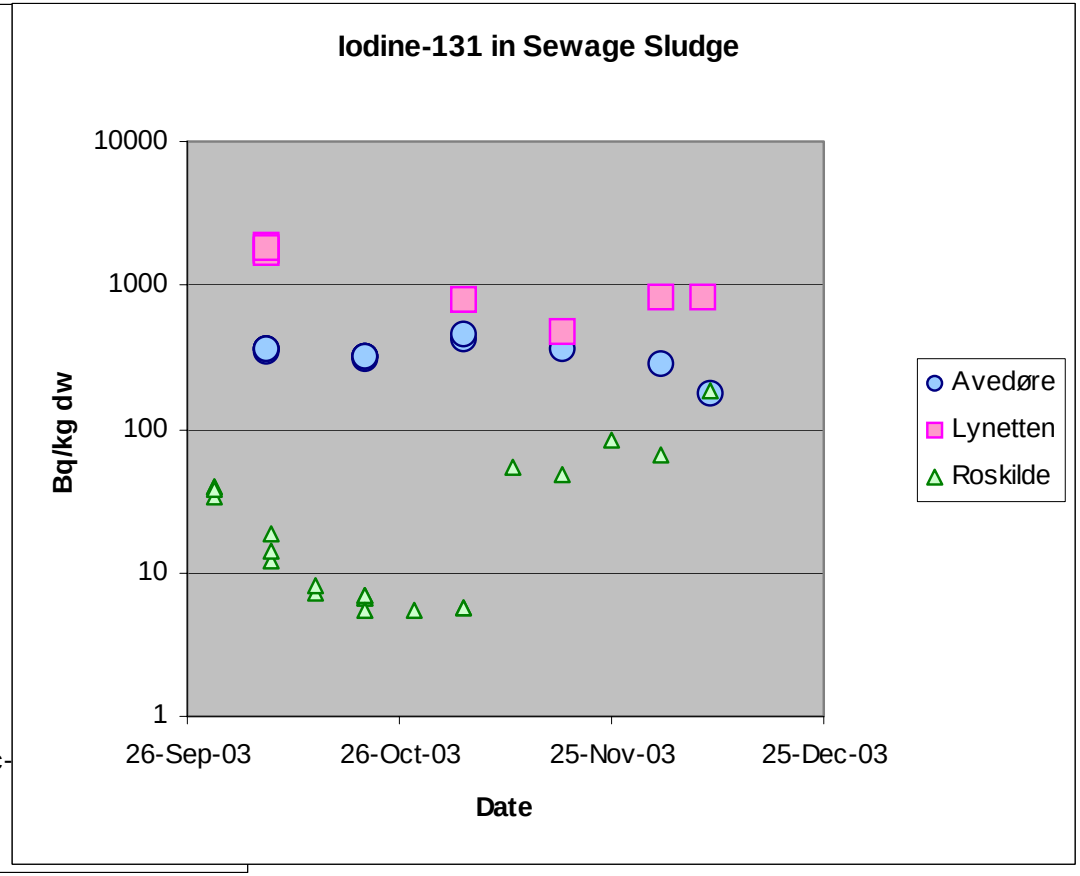
I-131 in Water and Sludge at other Danish WWTPs





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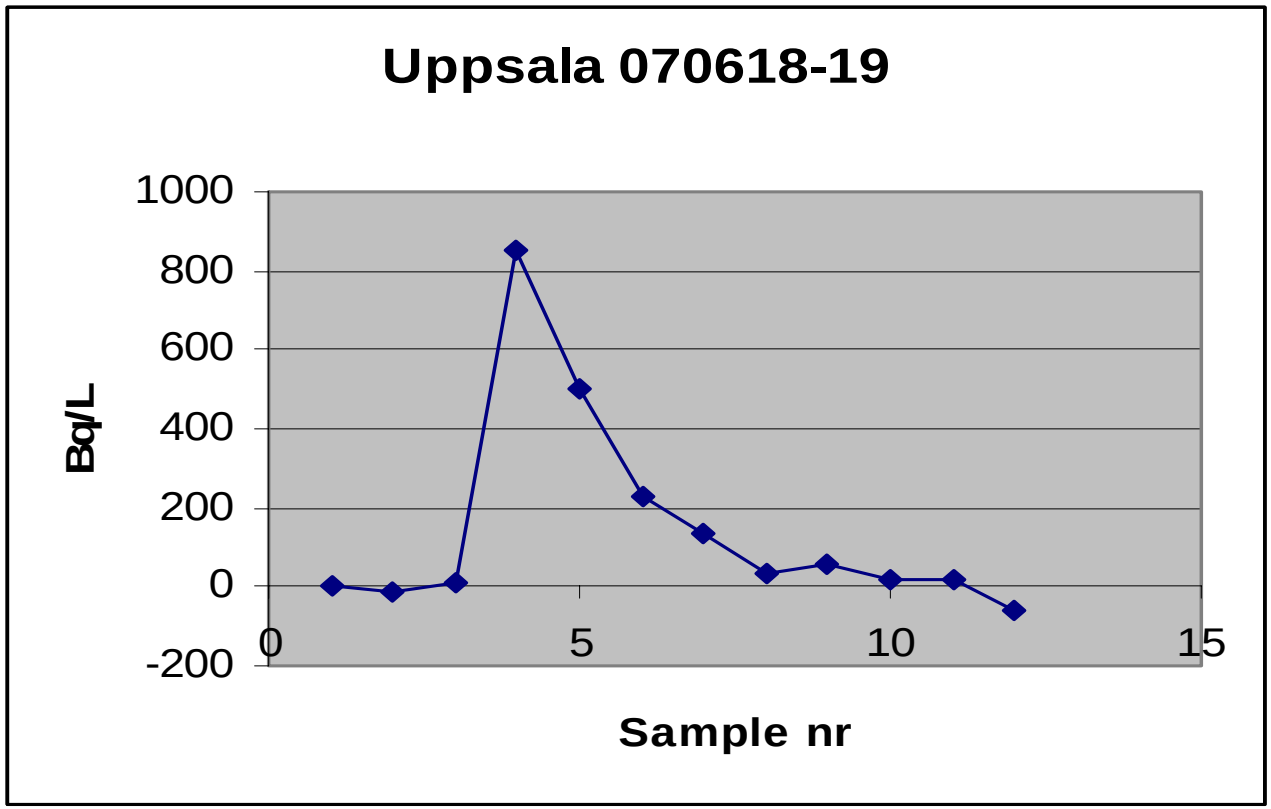
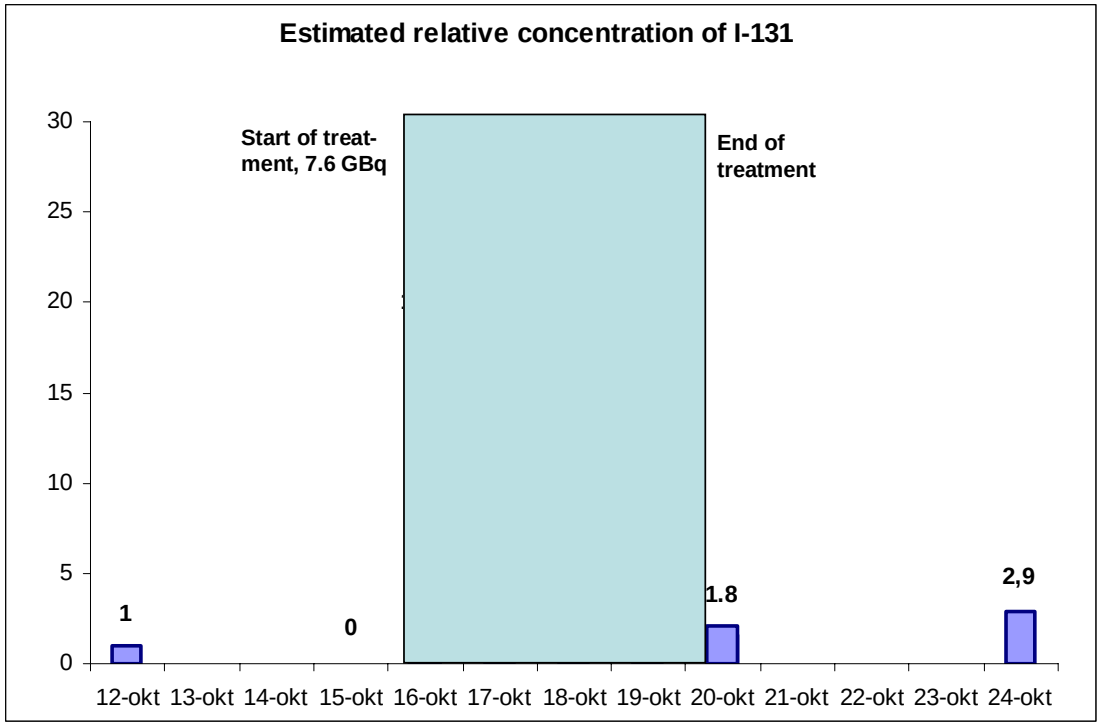
Activities of ^{131}I in Fucus vesiculosus near the outfall tunnel of the final effluent from the Helsinki wastewater treatment plant

Date	Bq kg-1 dry weight
23.11.1983	1.0
27. 9. 1985	0.4
30. 7 .1992	33
14. 9. 1995	210
8.7. 1999	57

Sample	Concentration of ^{131}I (Bq/kg)
Primary sludge, 18-oct 2007, wet weigth	204+/- 4
Primary sludge, 18 -oct 2007, dried over night at 105°C covered with folie	8120+/-230
Primary sludge, 18-oct 2007, dried over night uncovered	8560+/-730
Secondary sludge, 18-oct 2007 wet weight	185±2,5
Secondary sludge, 18-oct 2007 dried over night, covered with folie	13800±500
Secondary sludge, 18-oct 2007, dried over-night uncovered	16000±1360
Secondary sludge 22-oct 2007, wet weight	355±7,5
Secondary sludge 22-oct 2007, filtered	7980±152
Secondary sludge 22-oct 2007, filtrate	46±1,5
Secondary sludge, 23-oct 2007, wet weight	322±7,8
Secondary sludge, 23-oct 2007, filtered	6100±89
Secondary sludge, 23 -oct 2007, filtrate	37±1,9
Secondary sludge 24-oct 2007, wet weight	357±6,4
Secondary sludge, 24-oct 2007, filtered	6290±103
Secondary sludge, 24-oct 2007, filtrate	35±3,4
Primary Sludge, 9-jun 2005 wet weight	862±1,2
Primary sludge, 9-jun 2005 filtered	1945
Primary Sludge, 9-jun 2005 filtrate	217





Amounts of I-131 given at LSH : **7.6** **0.3 GBq** **0,7 GBq**

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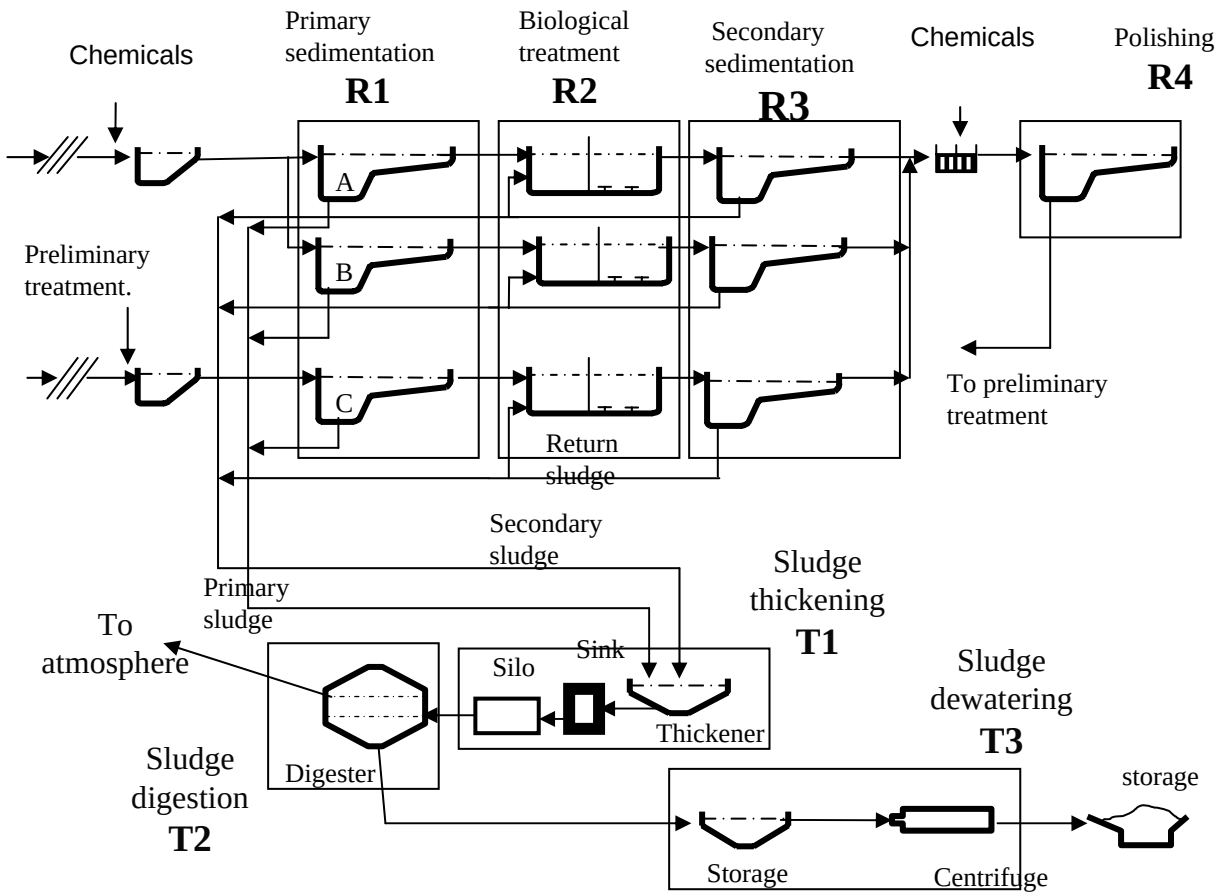

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Component → i / j process ↓	S_I	S_S	X_I	X_S	$X_{B,H}$	S_O	Process Rate, ρ_j [$ML^{-3}T^{-1}$]
Aerobic growth of heterotrophs		$-\frac{1}{Y_H}$			1	$\frac{1-Y_H}{Y_H}$	$\hat{\mu}_H\left(\frac{S_S}{K_S+S_S}\right)\left(\frac{S_O}{K_{O,H}+S_O}\right)X_{B,H}$
'Decay' of heterotrophs		1			-1		$b_H X_{B,H}$
'Hydrolysis' of entrapped organics		1		-1			$k_h \frac{X_S/X_{B,H}}{K_X+(X_S/X_{B,H})}\left[\left(\frac{S_O}{K_{O,H}+S_O}\right)+\eta_h\left(\frac{K_{O,H}}{K_{O,H}+S_O}\right)\right]X_{B,H}$

$$r_i = \sum_j \nu_{ij}\rho_j$$

Soluble inert organic [M(COD)L^{-3}]	Readily biodegradable Substrate [M(COD)L^{-3}]	Particulate inert organic [M(COD)L^{-3}]	Slowly biodegradable substrate [M(COD)L^{-3}]	Active hetrotrophic biomass [M(COD)L^{-3}]	Oxygen [M(COD)L^{-3}]

Monod-Herbert model

the Kd concept

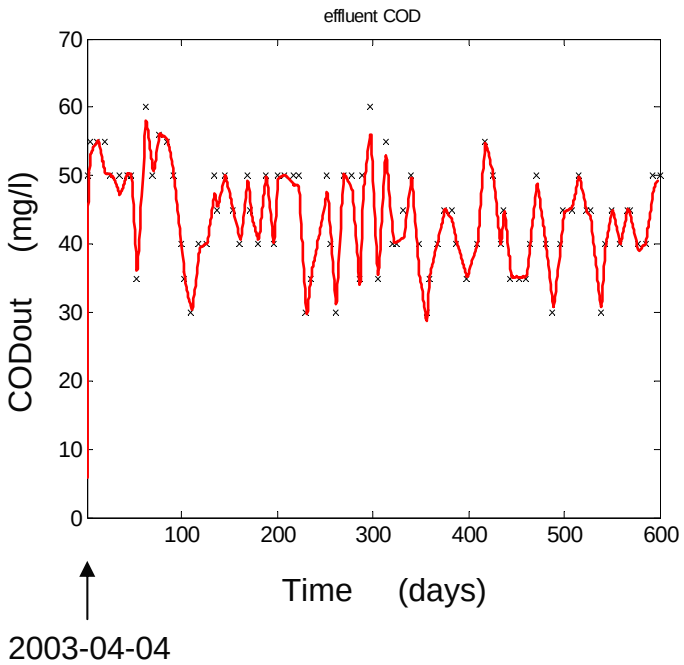
- The total inventory *(Atotal)* of radionuclides in a sewage basin will be:

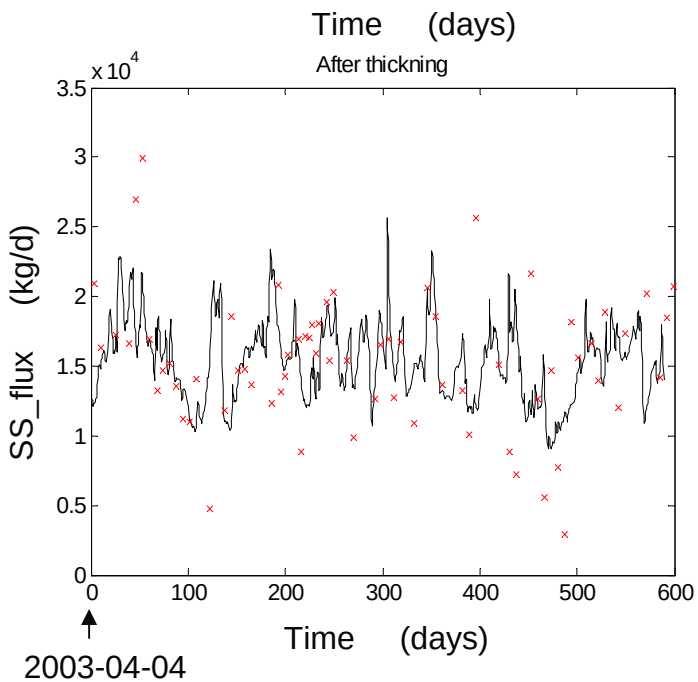
$$A_{total} = A_{solids} + A_{water} = Conc_{solids} * M_{solids} + Conc_{water} * V_{water}$$

- Diving by the water volume (*Vwater*) and introducing the Kd gives:

$$\frac{A_{total}}{V_{water}} = Conc_{water} * \left(Kd * SS + 1\right)$$

Modelling results





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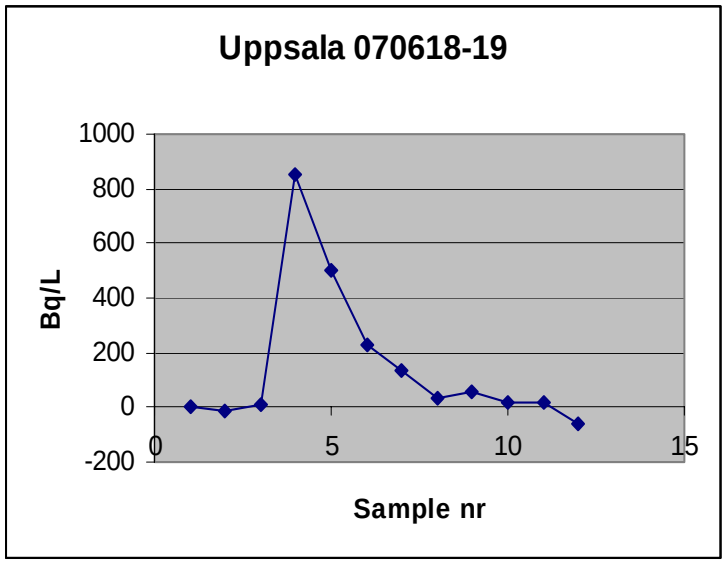
Measured Lu-177 at Uppsala plant



	Bq/kg
Inkommande vatten rör AB	11.5
Inkommande vatten rör C	16
Rötslam efter cent.	1540
Utgående till fyrisån	52.4

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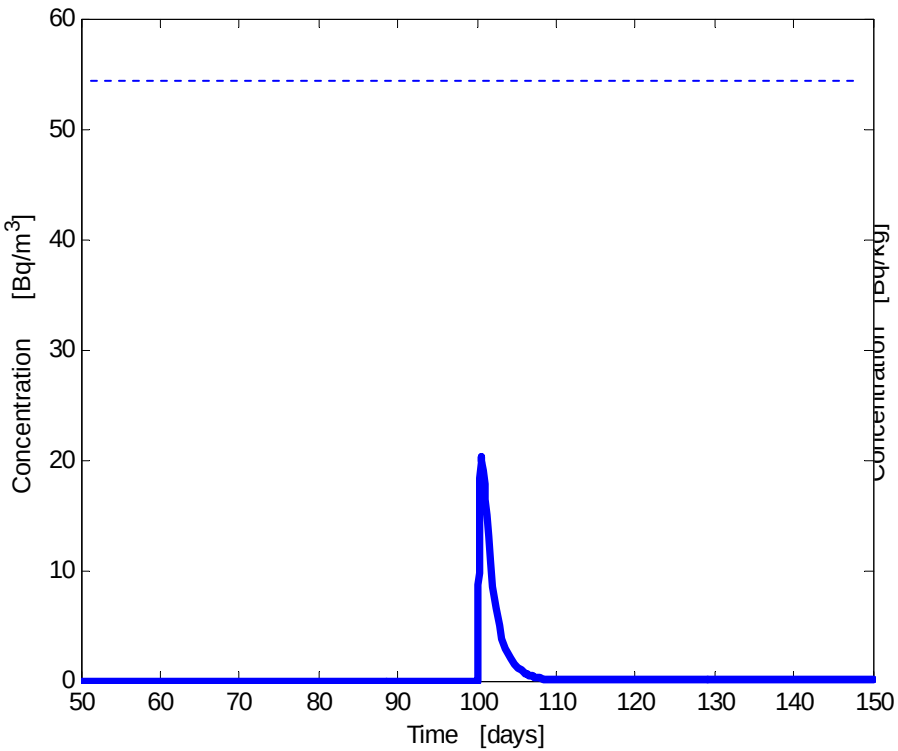
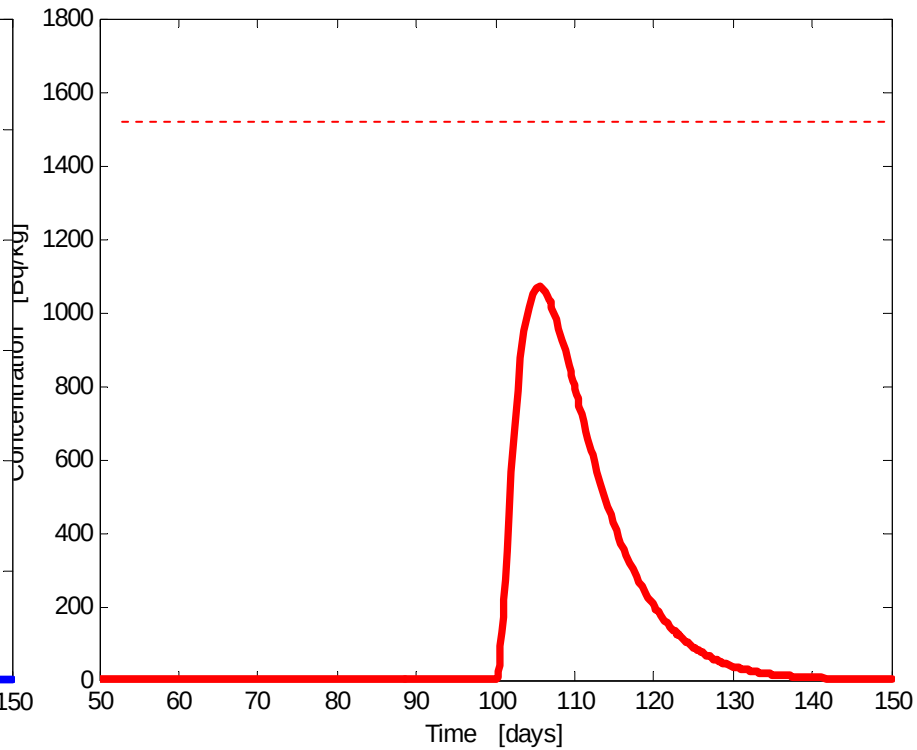
Modelling responses on the pulse release





K_d=0.001 m^3/g

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Water pathways

- Drinking water from the recipient
- Eats fish living in this acquifer
- No dilution in the final recipient assumed.
- Activity concentrations in water and fish multiplied by the consumption rates and DCF (ingestion)

Sludge pathways

- Sewage workers
- Farmers
- Inhalation and external radiation for sewage workers
- Inhalation, external radiation and ingestion of crops, meat and milk
- Activity concentrations in crops, meat and milk
- multiplied by the consumption rates and DCF (ingestion)

Dose factors for ingestion, inhalation (IAEA ,2001) and external exposure (Titley et al 2002)

Nuclide	D^j_{ing} Sv/Bq	D^j_{inh} Sv/Bq	D^j_{ex} Sv/a per Bq/m^3
I-131	2,2E-08	7,4E-09	7,1E-10
Tc-99m	2,2E-11	1,2E-11	2,6E-10

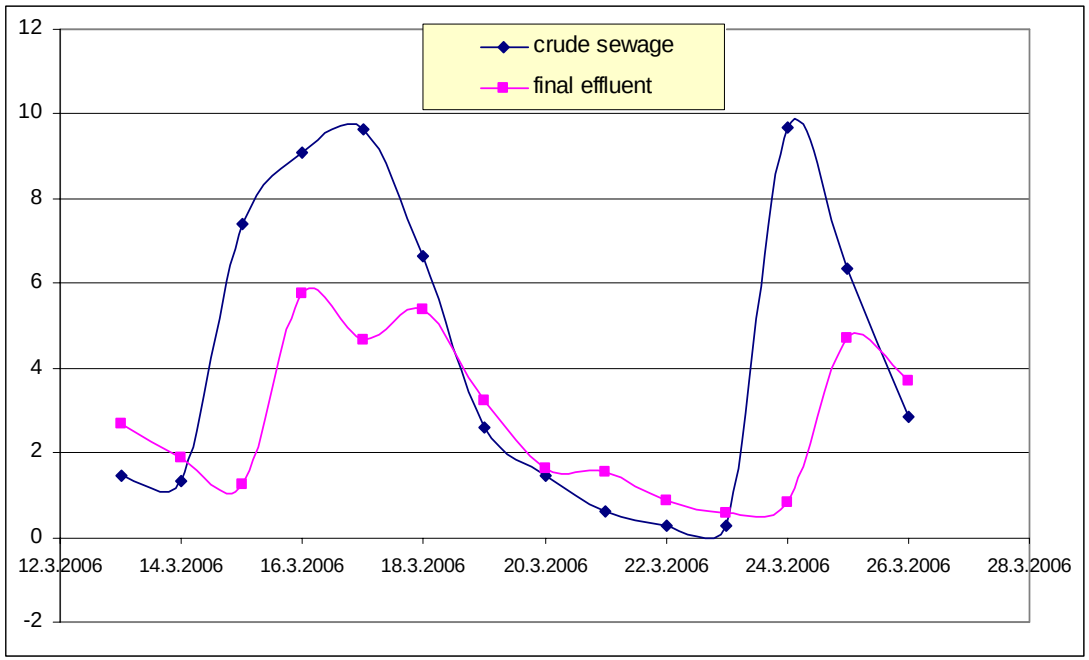
Extenal and inhalation annual dose rates to farmers

Sewage plant	Nuclide	$Dose_{ext,\ farmer}$ Sv/a	$DF_{inh,\ farmer}$ Sv/a	$Dose_{totalb}$ Sv/a
Kungsängverket, Sweden	Tc-99m	1,3E-13	4,2E-21	1,3E-13
	I-131	3,6E-09	3,2E-14	3,6E-09
Viikinmäki, Finland	Tc-99m	1,7E-13	6,4E-21	1,7E-13
	I-131	3,6E-09	3,2E-14	3,6E-09
VEAS Vestfjorden avlöpsselskap, Norway	Tc-99m	1,1E-13	4,2E-21	1,1E-13
	I-131	2,8E-09	2,4E-14	2,8E-09
Renseanlaeg Vest, Denmark	I-131	3,9E-09	3,4E-14	3,9E-09
Skólpa Klettagardar, Iceland	I-131	6,6E-09	5,8E-14	6,6E-09

Accidental situations

- Consider the dynamics in the variation of the activity concentrations in sewage water and sludge;
- More proper assessments of the time variations of the dose estimations;
- More proper identification of people affected;
- Scenarios…..???

^{131}I in sewage at Viikinmäki



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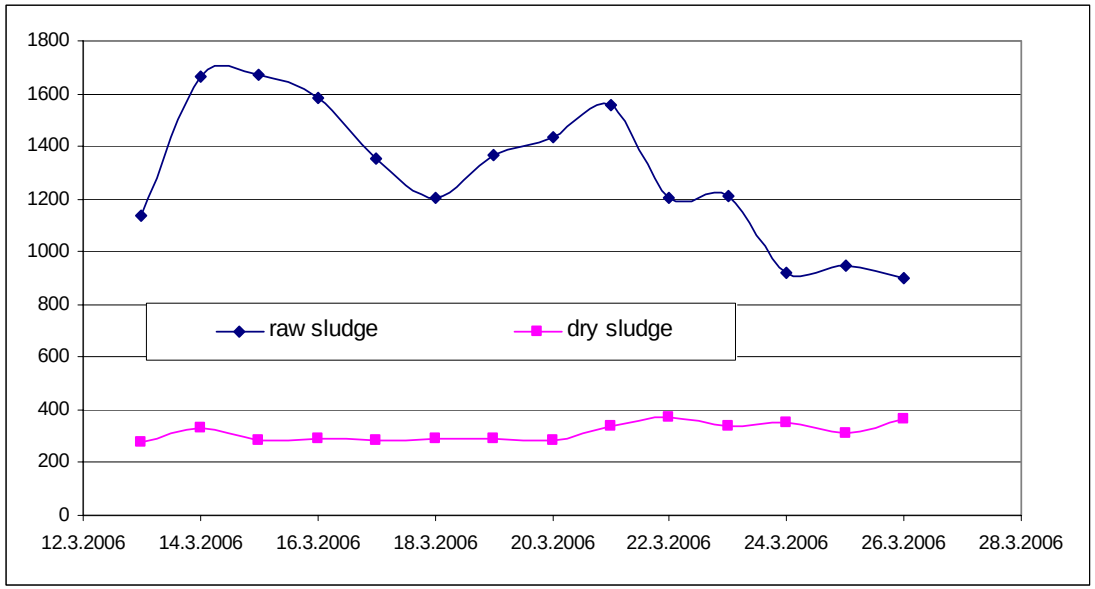
^{131}I in sludge at Viikinmäki



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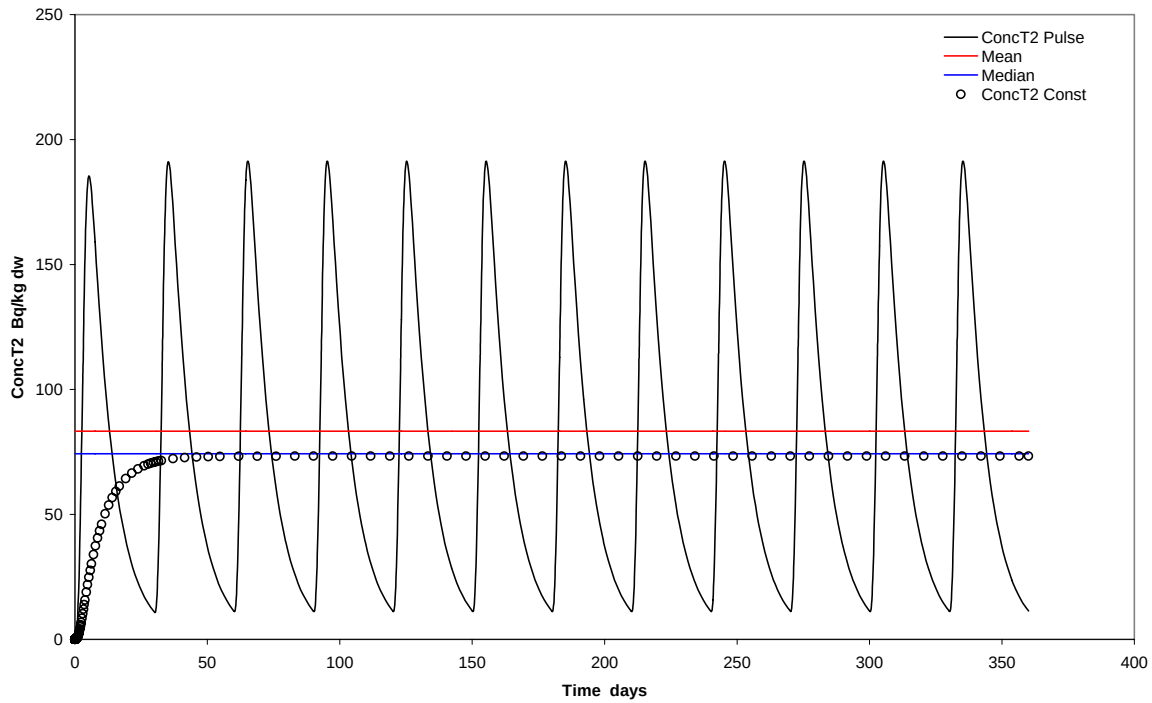
Dynamics of the releases



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Parametervalues for doses to workers

Correction coefficient	Units	Value
Density of the sewage sludge, ρ	kg/m^3	1000
Conversion factor between dry weight and wet weight, $Conv$	kg DW/kg WW	0.25
Fraction of the year during which exposure occurs, OF	dimensionless	0.228
Concentration of resuspended particles in air, RF	kg/m^3	1E-7