Radioactivity in produced water from Norwegian oil and gas installations - concentrations, bioavailability and doses to marine biota

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Outline

- Background – Radioactivity in produced water
- Radioactivity in produced water from NCS
- Project
  - WP1: Background and sources (IFE)
  - WP2: Speciation and mobility (IFE)
  - WP3: Bioavailability (NIVA)
  - WP4: Biological effects (NIFES)
  - WP5: Modelling (SINTEF)
  - WP6: Risk assessments IFE)
- Conclusion

Radioactivity in produced water

- ^{238}U and ^{232}Th present in the reservoir bedrock
- ^{226}Ra (daughter ^{238}U) and ^{228}Ra (daughter of ^{232}Th)
- Radium is dissolved in the formation water and transported through the production system
- Radium analogues: Ba, Ca, Sr
- Scale formation
- Scale inhibitors
- Radium not precipitated, follows produced water

Components in produced water

- Water and salts
- Hydrocarbons
- Organic acids
- Heavy metals and radionuclides
- Added chemicals
  - Scale inhibitors, corrosion inhibitors, biocides, flocculants, pH modifiers, etc….

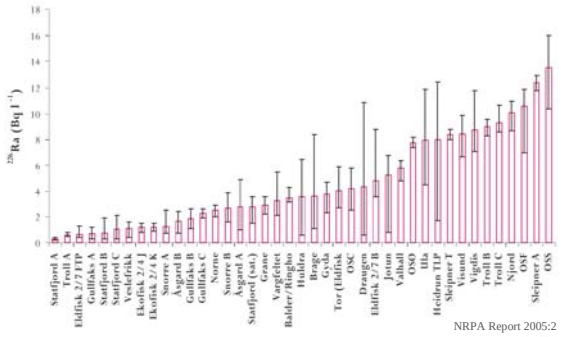
Discharge on the Norwegian Continental Shelf (GBq)

	^{226}Ra	^{228}Ra	^{210}Pb
2002	~ 300	-	-
2003*	~ 440 (3.3 Bq/l)	~ 380 (2.8 Bq/l)	< 92

Discharge volume in 2003: 135 Mm^3

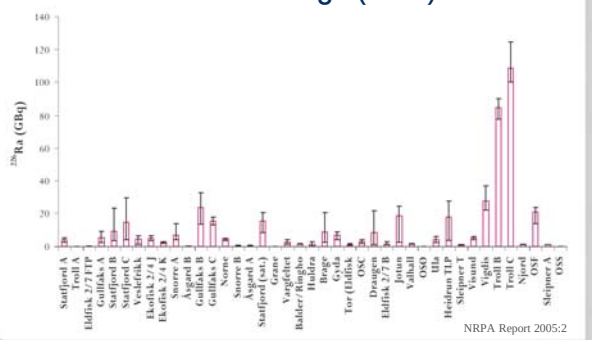
* NRPA Report 2005:2

^{226}Ra-discharge (2003)



NRPA Report 2005:2

^{226}Ra-discharge (2003)



NRPA Report 2005:2

^{226}Ra in the North Sea

- 1-2 mBq/l seawater
- (Troll B and C produced water: 9-10 Bq/l)

Questions raised

- What happens with discharged radionuclides?
- Does the addition of production chemicals effect the chemistry and biological uptake of discharged radionuclides?
- Are the effects of increased input of ^{226}Ra, ^{228}Ra observable?

The Project

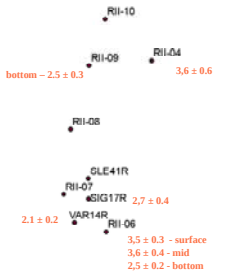
- WP1: Background and sources
- WP2: Speciation and mobility
- WP3: Bioavailability
- WP4: Biological effects
- WP5: Modelling with the DREAM model
- WP6: Risk assessments

WP1 - Background levels

- Seawater (10 samples; ^{226}Ra and ^{228}Ra)
- Sediment (32++ samples; ^{226}Ra, ^{228}Ra and ^{210}Pb)
- Sediment dwel. org. (9 locations; ^{226}Ra and ^{228}Ra)
- Fish (30 cod family fish; ^{226}Ra and ^{228}Ra)
  - Flesh, bone and skin

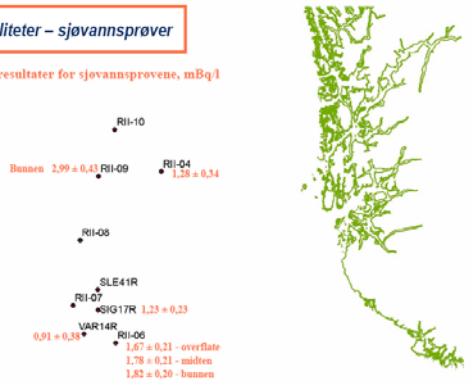
Location – seawater + bottom feeders

Results for seawater, mBq/l



Lokaliteter – sjøvannsprøver

^{226}Ra-resultater for sjøvannsprovene, mBq/l



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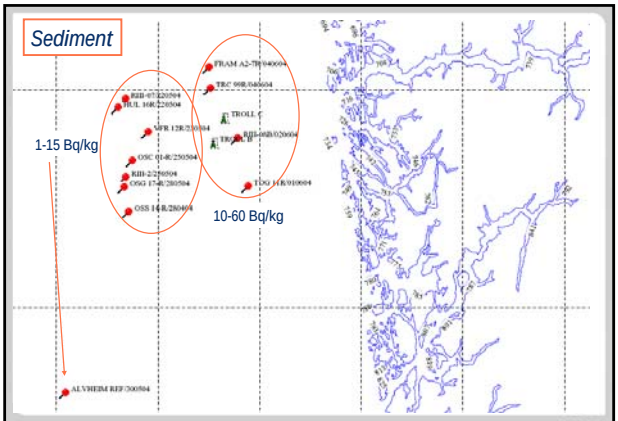
Sediment



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Data fra Karl Henrik Bryne i Statoil:

Sedimentkarakteristikker og dybde

Grain size composition (% dry weight)

SAMPLE	clay	silt	pelite	very fine sand	fine sand	medium sand	coarse sand	very coarse sand	total sand	pebbles	cobbles	gravel
R III-02	N/A	N/A	3,53%	4,69%	78,56%	12,56%	0,63%	0,02%	96,44%	0,06%	0,00%	0,06%
R III-06	N/A	N/A	96,84%	1,53%	0,69%	0,78%	0,13%	0,03%	3,14%	0,03%	0,00%	0,03%
R III-07	N/A	N/A	3,03%	13,00%	73,36%	3,35%	2,30%	2,40%	94,41%	2,56%	0,00%	2,56%
R III-08B	N/A	N/A	98,22%	0,70%	0,48%	0,38%	0,12%	0,10%	1,78%	0,00%	0,00%	0,00%
VFR 12R	N/A	N/A	4,17%	21,39%	72,51%	1,35%	0,26%	0,23%	95,73%	0,10%	0,00%	0,10%
HUL 16R	N/A	N/A	1,70%	1,13%	43,84%	36,29%	9,60%	4,53%	95,40%	2,90%	0,00%	2,90%
OSS 14R	N/A	N/A	1,42%	0,86%	31,58%	59,24%	6,47%	0,36%	98,52%	0,06%	0,00%	0,06%
OSC 01R	N/A	N/A	1,66%	2,20%	61,08%	25,35%	5,13%	4,13%	97,89%	0,45%	0,00%	0,45%
OSE 14R	N/A	N/A	4,48%	17,75%	64,83%	4,44%	2,14%	3,57%	92,72%	2,80%	0,00%	2,80%
OS-G 17R	N/A	N/A	2,59%	5,93%	89,17%	2,17%	0,13%	0,00%	97,41%	0,00%	0,00%	0,00%
TOG 11R	N/A	N/A	96,22%	1,01%	0,88%	1,04%	0,78%	0,08%	3,78%	0,00%	0,00%	0,00%
TRC 99R	N/A	N/A	98,38%	0,36%	0,27%	0,36%	0,52%	0,11%	1,62%	0,00%	0,00%	0,00%
FRAM A2-7R	N/A	N/A	99,14%	0,31%	0,20%	0,23%	0,04%	0,08%	0,86%	0,00%	0,00%	0,00%
average value			39,33%	5,45%	39,80%	11,35%	2,17%	1,20%	59,98%	0,69%	0,00%	0,69%
minimum value			1,42%	0,31%	0,20%	0,23%	0,04%	0,00%	0,86%	0,00%	0,00%	0,00%
maximum value			99,14%	21,39%	89,17%	59,24%	9,60%	4,53%	98,52%	2,90%	0,00%	2,90%
st. dev			48,09%	7,21%	35,38%	18,24%	3,03%	1,77%	47,54%	1,19%	0,00%	1,19%

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Fisk

Prøve	^{226}Ra (Bq/kg fv)	^{228}Ra (Bq/kg fv)
Torsk, Barentshavet:		
Kjøtt	< 0,3	< 0,4
Skinn	0,5	< 0,6
Bein	< 0,5	< 1,0
Skrei, Lofoten:		
Kjøtt	< 0,2	< 0,3
Bein	< 0,5	< 0,6
Sei, Nordsjøen		
Kjøtt	< 0,2	< 0,3
Skinn	0,3	0,3
Bein	< 0,5	< 0,8

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WP 2 – Speciation and mobility

- Determine the levels and distribution of relevant radionuclides in produced water
- Study mobility and sedimentation mechanisms of the radionuclides

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Troll B og C (2004)

- Produced water: 23,2 Mm^3
- ^{226}Ra: 209 GBq
- Ba: 4660 tonnes
- Scale inhibitor: 306 tonnes (SI 4470)

- ^{226}Ra: 9 Bq/l
- Ba: 200 ppm
- SI: 13 ppm
- Sulfate: 0

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Troll B og C

- ^{210}Po: Meget lave nivåer (omr. mBq/l)
- ^{210}Pb: Meget lave nivåer (omr. mBq/l)
- Formasjonsvannet har reduserende betingelser → sulfidinnholdet fører til at Pb felles som PbS og transporteres ikke med produsert vann

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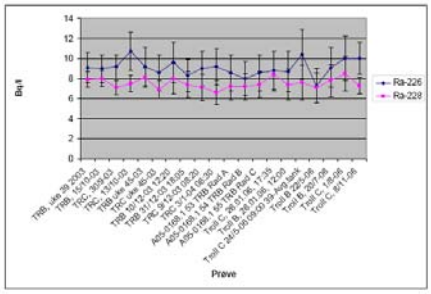
^{226}Ra og ^{228}Ra – Troll B og C



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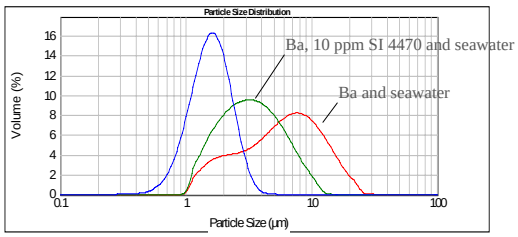
$BaSO_4$ particle size distribution



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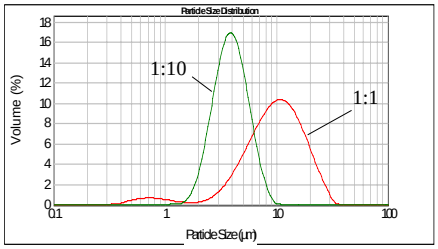
Mixing of "produced water" with seawater



SI 4470 concentration: 10 ppm

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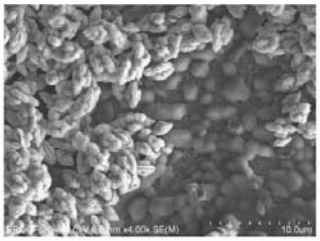
Partikkelmorfologi



200 ppm Ba, 10 ppm SI 4470 pluss sjøvann

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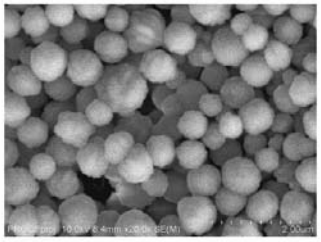
Partikkelmorfologi



200 ppm Ba, 200 ppm SI 4470 pluss sjøvann

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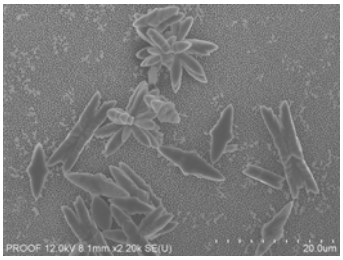
Morphology – 1:1 dilution



SI 4470 concentration 10 ppm

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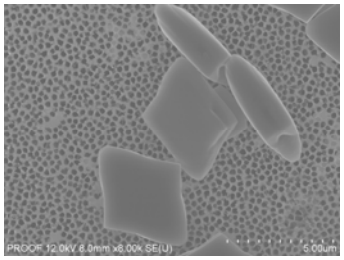
Morphology – 1:10 dilution



SI 4470 concentration 10 ppm

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Blanding av kunstig produsert vann med sjøvann

- Kunstig Troll B/C vann lages ved å tilsette dest. vann Na, K, Mg, Ca, Ba, ^{133}Ba, Sr, Cl, bikarbonat og SI
- Oppvarmes til 60 ^{o}C
- Tynnes ut med kaldt sjøvann: 1:1, 1:10 og 1:100

- 1:1: 67±4% Ba partikkelbundet
- 1:10: 41±2% Ba partikkelbundet
- 1:100: 0,20±0,02% Ba partikkelbundet

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Forsøk på Troll C – partikkelbundet Ra

Bland. forhold	Prøve A	Prøve B	Prøve C
Ingen	0,7 ± 0,1 %	0,7 ± 0,1 %	0,6 ± 0,1 %
1:1	20 ± 4 %	100 ± 20 %	41 ± 8 %
1:10	73 ± 14 %	51 ± 10 %	7 ± 2 %
1:100	2,9 ± 0,8 %	1,4 ± 0,5 %	2,9 ± 0,8 %

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Hva skjer når produsert vann blandes med sjøvann?

- Grunnet høy kons. av Ba, vil $BaSO_4$ felles ut og ta med seg Ra
- Utfellingen, krystalldannelsen, vil avhenge av hvor mye SI som er tilstede og hvor raskt produsert vann tynnes ut. Treg fortynning og lav SI-kons. fører til at det dannes større $BaSO_4$-partikler
- Rask fortynning fører til dannelse av små $BaSO_4$-partikler
- $BaSO_4$ som først er dannet, løses trolig ikke opp, men partiklene blir mindre

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Konklusjon

- Produsert vann fra norske installasjoner har meget lavt innhold av Pb-210 og Po-210 (reduserende betingelser)
- Ra i produsert vann fra Troll B og C finnes på løst form (<0,45 µm)
- Radium som slippes ut med produsert vann feller ut med $BaSO_4$, men partiklene som dannes er små
- Fortynning av produsert vann med sjøvann fører til at $Ba(Ra)SO_4$-partiklene blir enda mindre

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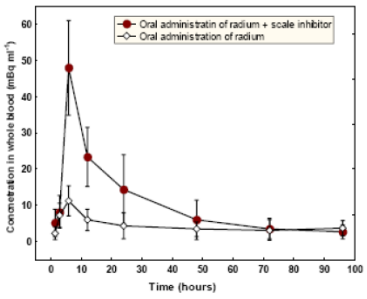
WP 3: Bioavailability

- Bioavailability of radium to fish
  - Dietary
  - Aquatic
- Bioavailability of radium to sediment-dwelling organisms

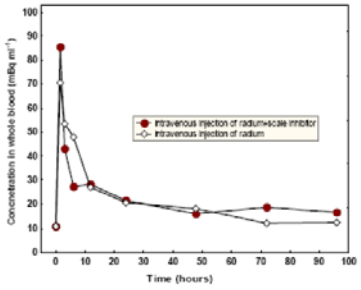
Dietary bioavailability

- Single oral or intravenous (i.v.) administration of Ra or Ra + SI 4470
- 36 fish in each group (4 groups)
- Sampling of blood from four fish per sampling point at 0, 1.5, 3, 6, 12, 24, 48, 72, 96 hours
- i.v. injection AUC is reference: 100% bioavailability
- Relative AUC oral/i.v. versus dose oral/i.v. = bioavailability (x%)

Results, oral



Results, i. v. injection



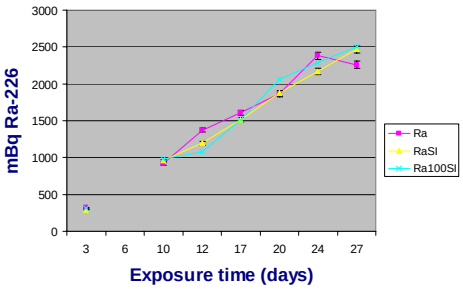
Results - Oral

- Food to blood bioavailability
  - Ra: 12%
  - Ra + SI: 60%
- Presence of SI 4470 increases the bioavailability of dietary Ra

Aquatic accumulation

- 4 groups, triplicate aquaria
  - Ra (10 Bq/l)
  - Ra + 0.13 mg/l SI 4470
  - Ra + 13 mg/l SI 4470
  - Control
- 27 days exposure, water change regularly (3-5 days)
- Samples taken before each water change:
  - Fish: skin, blood, liver, gills, kidney, carcass
  - Water
  - DGTs

MnO_2-DGTs



Results, aquatic uptake

- Blood
  - No differences or trends
- Liver
  - No differences
- Kidney
  - No differences or trends
- Gills
  - Increase in the presence of SI
- Skin
  - Increase in the presence of SI

Biotilgjengelighet og effekter på sedimentlevende organismer

- Sediment fra oslofjorden deles i fire grupper, 4 paralleller i hver gruppe

  1: Kun sediment
  2: Sediment + 0,5 Bq/g ^{226}Ra
  3: Sediment + 0,5 Bq/g ^{226}Ra + inhibitor
  4: Sediment + 5 Bq/g ^{226}Ra + inhibitor

- Inhibitor fører ikke til økt opptakt av Ra i mark
- ingen effekter påantioksidant-forsvaret hos *Hediste diversicolor*

Conceptual model for doses to biota

Uncertainty *Below background*	< 10 µGy/y- 40 µGy/y
Radiation well-being *Natural background*	40 µGy/y- 5 mGy/y
Physiological masking *Minor effects on individual*	5 mGy/y- 50 mGy/y
Ecological masking *Effects on population level*	50 mGy/y- 4 Gy/y
Obvious action *Reduction, elimination*	4 Gy/y – 3000 Gy/y

Polikarpov (1998)

Intern dose, Ra-226

Prøve	Doserate [µGy/t]	Dose pr år [µGy]
Torsk, Barentshavet	$1{,}1 \cdot 10^{-2}$	96
Torsk, Lofoten	$<3{,}6 \cdot 10^{-3}$	<32
Sei, Nordsjøen	$<3{,}6 \cdot 10^{-3}$	<32

Intern dose, Ra-228

Prøve	Doserate [µGy/t]	Dose pr år [µGy]
Torsk, Barentshavet	$<2{,}4 \cdot 10^{-3}$	<21
Torsk, Lofoten	$<1{,}6 \cdot 10^{-3}$	<14
Sei, Nordsjøen	$<0{,}8 \cdot 10^{-3}$	<7

Ekstern dose til fisk

- Beregning av ekstern doserate fra aktivitetskonsentrasjon i sjøvann

^{226}Ra i sjøvann	Ekstern doserate [µGy/h]	Ekstern årsdose [µGy/y]
Bakgrunnsnivå 2 mBq/L	$4{,}7 \cdot 10^{-6}$	0,04
Troll prod. vann 1:100 fortynning	$9{,}3 \cdot 10^{-5}$	0,8
Troll prod.vann 10 Bq/L	$9{,}3 \cdot 10^{-3}$	81,5

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Absorbed dose calculations

- Results from initial calculations
- External dose to fish, only ^{226}Ra included

^{226}Ra i sea-water	External dose [µGy/h]	External dose [µGy/y]
Background level 2 mBq/L	$4.7 \cdot 10^{-6}$	0.04
Troll prod.water 1:100 dilution	$9.3 \cdot 10^{-5}$	0.8
Troll prod.water 10 Bq/L	$9.3 \cdot 10^{-3}$	81.5

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Absorbed dose calculations (cont)

- Internal dose
- Final calculation will be based on input from uptake- and bioavailability studies
- Example using 2 Bq/kg (IAEA, high-end of observations)

^{226}Ra Atlantic cod	Internal dose [µGy/h]	Internal dose [µGy/y]
Background level Tissue < 0.1 Bq/kg	$1.8 \cdot 10^{-3}$	16
Example 2 Bq/kg	$3.6 \cdot 10^{-2}$	315

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Konklusjon

- Radium som slippes ut med produsert vann feller ut med $BaSO_4$, men partikklene som dannes er meget små
- Radiumutslippet med produsert vann har ingen observerbar effekt på hverken miljø eller biota

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