Iodine-131 and other medical radioisotopes in *Fucus vesiculosus* in the marine environment around Tromsøya and in sewage from the University Hospital of North Norway in Tromsø



#### Referanse

Gwynn JP, Kiel Jensen L. lodine-131 and other medical radioisotopes in *Fucus vesiculosus* in the marine environment around Tromsøya and in sewage from the University Hospital of North Norway in Tromsø.

Teknisk dokument nr. 15. Østerås: Direktoratet for strålevern og atomsikkerhet, 2019.

#### Reference

Gwynn JP, Kiel Jensen L. lodine-131 and other medical radioisotopes in Fucus vesiculosus in the marine environment around Tromsøya and in sewage from the University Hospital of North Norway in Tromsø.

Technical Document no. 15. Østerås: Norwegian Radiation and Nuclear Safety Authority, 2019.

#### Key words

lodine-131. Medical radioisotopes. Fucus. Sewage. Dose

#### **Abstract**

It is possible to establish a direct link between the use of medical radioisotopes at the University Hospital of North Norway and observations of activity concentrations of the same radioisotopes in sewage and in the case of iodine-131 (I-131) in the brown seaweed *Fucus vesiculosus* in the marine environment around Tromsø. This study estimates biological half lifes of I-131 in *F. vesiculosus*, excretion rates of medical radioisotopes from patients as well as biota and human dose exposures.

Prosjektleder: Gwynn JP & Kiel Jensen L

Godkjent:

Per Strand

Avdelingsdirektør, avdeling atomsikkerhet og miljø

Publisert Sider 09.12.2019

36

DSA,

Postboks 55, No-1332 Østerås.

Norge.

Telefon Faks Email 67 16 25 00 67 14 74 07 dsa@dsa.no dsa.no

ISSN 2387-5240

Iodine-131 and other medical radioisotopes in Fucus vesiculosus in the marine environment around Tromsøya and in sewage from the University Hospital of North Norway in Tromsø

Justin P. Gwynn & Louise Kiel Jensen

fra Direktoratet for strålevern og atomsikkerhet (DSA)

Østerås, 2019, Norway

### Content

Introduction		5
Backgrou	nd to study	5
1	University Hospital of North Norway (UNN)	8
2	Breivika wastewater treatment plant (WTP)	9
2.1	Sampling and analysis of <i>F. vesiculosus</i>	10
2.2	Results for F. vesiculosus	10
2.3	Effective and biological half lifes of I-131 in F. vesiculosus	18
2.4	Dose assessment for F. vesiculosus and human pathways	21
2.5	Sampling and analysis of sewage samples	22
2.6	Results for I-131 in sewage samples	22
2.7	Results for Tc-99m in sewage samples	27
2.8	Results for other radioisotopes in sewage samples	30
3	Consideration of possible exposures and doses to workers at Breivika WTP	31
4	Conclusions	33
Acknowledge	ement	34
References	35	

#### Introduction

The use of radioisotopes in the field of medical diagnostic radiography and radiotherapy is a well established and growing sector. According to the World Nuclear Association, more than 50 different activation or fission products have been identified for use in certain medical procedures and are produced either in nuclear reactors or by cyclotrons. Radioisotopes identified for medical procedures are mainly gamma and/or beta emitters, with typically short half-lifes ranging between minutes and days. Two of the most commonly used medical radioisotopes are Tc-99m (half-life 6 hours), used for diagnostic radiography, and I-131 (half-life 8 days), which is widely used for the radiotherapy of thyroid cancer. Medical facilities are normally authorised to discharge radioisotopes to sewage systems by national authorities. Discharges typically result from the excretion of radioisotopes in urine and faeces from patients undergoing diagnosis or treatment while they are admitted to or present at the medical facility. When receiving high activity doses, it is often necessary for patients to remain at the medical facility until the activity of the radioisotope within the body has decayed or reduced below a certain threshold. Discharges from patients receiving medical radioisotopes may also occur to local sewage systems when patients are allowed to return home following their procedure. The activity of medical radioisotopes in use at an authorised medical facility typically increases with increasing population size. However, the activity discharged from a medical facility can be reduced though the use of delay tanks that capture waste from patients undergoing radiotherapy. This allows for the further physical decay of radioisotopes before the contents of the delay tanks are emptied into the local sewage system. However, the use of delay tanks by medical facilities varies between and within different countries. Medical radioisotopes discharged to the sewage system typically must pass through wastewater treatment plants (WTP) before being discharged to rivers or directly to the marine environment. The type of WTP (primary, secondary or tertiary) and the time taken for any radioisotopes to pass through the WTP can impact the activity of the radioisotope that is finally discharged into the receiving water. Residence times for medical radioisotopes in sewage processed by secondary and tertiary WTPs would be expected to be longer than for primary WTPs.

#### **Background to study**

While undertaking monitoring of the marine environment around the Grøtsund port facility at Tønsvika in 2016, Iodine-131 (I-131) was observed (0.7 to 1.5 Bq/kg f.w.) in 6 samples of the brown seaweed *Fucus vesiculosus*. The only known local source of this radionuclide is the use of I-131 in the radiotherapy treatment of patients at the University Hospital of North Norway (UNN) in Tromsø. The discharge point for effluent from UNN is approximately 10 km from the Grøtsund port facility (Fig.1).

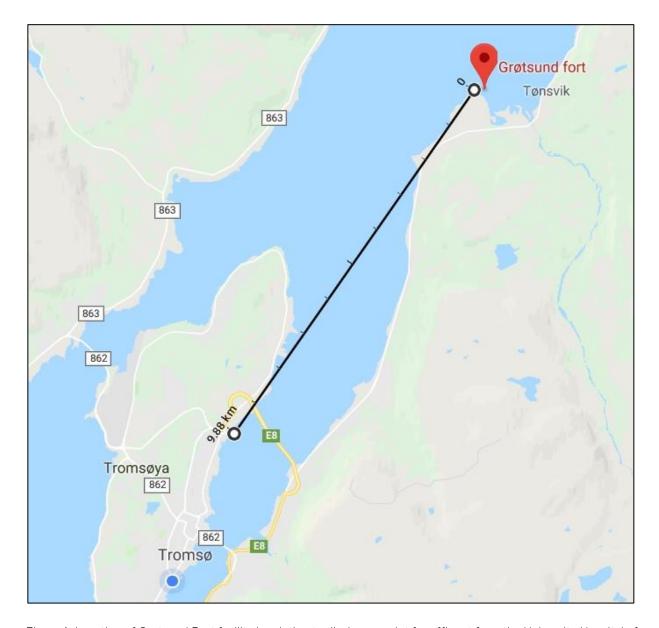


Figure 1. Location of Grøtsund Port facility in relation to discharge point for effluent from the University Hospital of North Norway in Tromsøsundet.

In order to establish whether the source of the I-131 observed in these seaweed samples was from UNN and to determine the extent and degree of impact of any such releases, further sampling of *F. vesiculosis* was carried out at various locations around Tromsøya. In addition, samples of influent, effluent and solid waste were collected at the Breivika WTP which receives sewage directly from UNN.

#### Fucus vesiculosus

F. vesiculosus is an edible and ubiquitous brown seaweed found in sheltered intertidal zones along the coast and shore line of Northern Norway (Fig.2) and is often used as a sentinel bio-indicator for certain contaminants. For example, F. vesiculosus and other brown seaweeds are known to accumulate iodine isotopes and Tc-99 (e.g. Druehl et al., 1988; Cooper et al., 1998; Brown et al., 1999). Bioaccumulation of iodine by Fucus spp. has been shown to be an active process, which can be influenced by metabolic effects (Klemperer, 1957). Recommended biological concentration factors (BCF) for Technetium and Iodine are  $3 \times 10^4$  and  $1 \times 10^4$ , respectively (IAEA, 2004). F. vesiculosus is sold as a nutritional supplement, but no commercial harvesting or aquaculture of this seaweed occurs in the area.



Figure 2. Fucus vesiculosus in the intertidal zone at one of the sampling stations.

### 1 University Hospital of North Norway (UNN)

UNN has permission for the use and discharge of a range of different radioisotopes for the purposes of radiotherapy and diagnostic radiography (Table 1). For regulatory purposes, the amount of each radioisotope discharged to the sewer for any given year through bodily wastes is calculated from the amount used in combination with recommended excretion factors for the chemical form of the radioisotope (IPEM, 2018). I-131 is administered at UNN as NaI in capsule form and it is assumed that 100% of the administered dose is excreted to the sewage system. However, according to the ICRP (2004) only 84% to 90% of I-131 administered for the treatment of thyroid carcinomas is discharged to sewage systems. At the time of the monitoring in 2017, UNN did not have any decay tanks for initial handling of sewage from patients undergoing radiotherapy or diagnostic radiography. A delay tank has been installed as part of the new PET centre at UNN, but waste from patients receiving I-131 is not collected and is still discharged directly to the sewage system. Patients receiving I-131 (3.7 to 6 GBg) at UNN are typically isolated for 3 days until their thyroid measurements give dose rates below 30 μSv/h. Urinary excretion is the predominant route for loss of iodine (35% to 75% in 24 hours), although there is some faecal excretion as well (Mettler and Guiberteau, 2012). In the period 2013 to 2017, between 26 and 41 patients were treated at UNN for radiotherapy with I-131 per year. In 2017, the total activity of I-131 discharged from UNN was reported as 0.1 TBq (Table 1), compared to an estimated total of 1.3 TBq for the whole of Norway. Patients receiving radioisotopes for diagnostic radiography (e.g. Tc-99m) are not isolated and may leave UNN before any or complete excretion of such radioisotopes into the local sewage system. UNN has previous carried out assessments of doses to sewage workers at the Breivika WTP and the ambient exposure at Breivika harbour. A sewage worker carried a dosimeter for 16 working days and recorded an average dose of 0.033  $\mu$ Sv/hr, equivalent to 67  $\mu$ Sv/a. The dosimeter placed in Breivika harbour for 44 days recorded an average dose of 0.085  $\mu$ Sv/hr, equivalent to 170  $\mu$ Sv/a.

Table 1. Discharge limits and discharges to sewage system for radioisotopes used by UNN

Radioisotope	Half-life	Discharge limit GBq/a <sup>1</sup>	Total discharge for 2017 (GBq)²
Tc-99m	6 hours	300	176
I-131	8 days	200	106
I-123	13.2 hours	10	5.6
Cr-51	27.7 days	1	0.17
Se-75	120 days	0.01	0.002
F-18	110 minutes	50	36
Ra-223	11.4 days	0.20	0.06
In-111	2.8 days	7	0
Sm-153	46.3 hours	15	0
Sr-89	50.6 days	1.05	0

<sup>1 -</sup> As authorised by the Norwegian Radiation and Nuclear Safety Authority. 2- as reported by UNN.

### 2 Breivika wastewater treatment plant (WTP)

The Breivika WTP is a primary treatment facility that filters solid material larger than 350 µm from incoming influent, before discharging effluent directly to Tromsøysundet. The Breivika WTP is designed to handle waste from 18700 person equivalents at flow rates up to 230 l/s. In periods when flow rates exceed capacity or when the WTP is offline for routine cleaning or maintenance, unfiltered influent can be discharged. The Breivika WTP receives domestic influent as well as influent from the University of Tromsø (UiT) and UNN. UIT only has permission for discharges to the sewage system for H-3 and C-14 as well as for I-125 to air. For the period 2007 to 2013, the Breivika WTP processed up to 3.3 million m<sup>3</sup> of influent per year and generated 500 to 700 tonnes of solid waste per year. Solid waste is collected in containers and then transported to a waste disposal site at Stormoen, Balsfjord for windrow composting for around 5 years. The Breivika WTP is fully automated but workers are required to undertake routine cleaning and periodic maintenance of the facility. The discharge point for effluent from the Breivika WTP is approximately 140 m from the shore at a depth of 17.4 m (Fig.3). Discharges of effluent would be expected to be fresher and warmer (i.e. positively buoyant) than the receiving sea water, so that any discharged I-131 or other radioisotopes should be dispersed within surface layers of Tromsøysundet. The maximum tidal range in Tromsøysundet is around 3 m with northward and southward water current flow rates in the area around the discharge point up to 0.5 m/s, although currents in other parts of Tromsøysundet can reach up to 1.5 m/s (Audunson & Næser, 1975; Statens Kartverk, 2008).

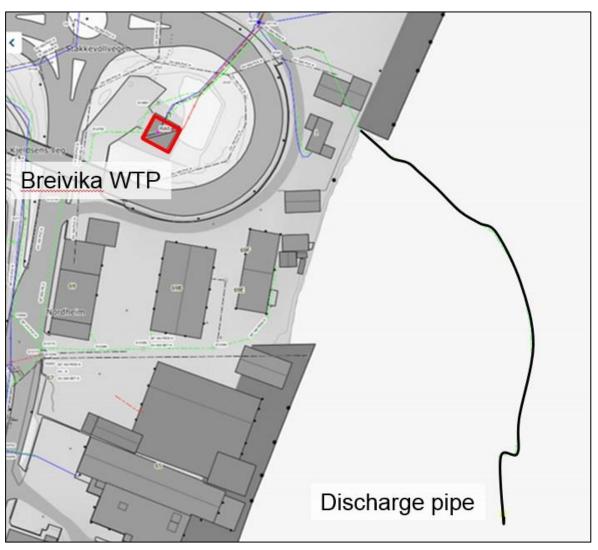


Figure 3. Position of offshore discharge pipe in relation to location of Breivika WTP.

#### 2.1 Sampling and analysis of F. vesiculosus

All seaweed samples were collected and analysed as bulk samples unless stated otherwise. All samples were checked for any contaminating material before analysis. Unless stated otherwise all seaweed samples were analysed without drying and immediately upon return to the laboratory. Where samples were dried prior to analysis (at 60 °C overnight), dry weight (d.w.) results have been converted back to fresh weight using either sample specific d.w./f.w. ratios or an average ratio of 0.25. Fresh weight samples were chopped, while dried samples were homogenised in a stainless steel blender. All samples were packed into plastic counting geometries and were analysed for at least 1 hour on HPGe detectors. All results were decay corrected to the sampling time and date and are given with associated uncertainties to 1 sigma.

#### 2.2 Results for F. vesiculosus

Initial sampling of *F. vesiculosus* was carried out in August 2016 at three different stations in the area around the discharge point of effluent from UNN. (Fig.4). Samples of *F. vesiculous* were collected on days before radiotherapy treatment of a patient with 4 GBq of I-131 at UNN and at set times afterwards. The last previous patient admitted for radiotherapy with I-131 at UNN was 20 days earlier.

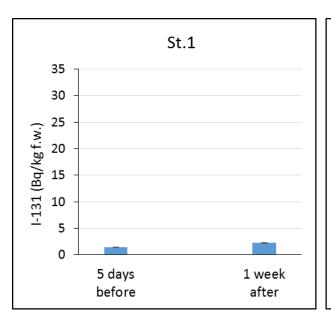


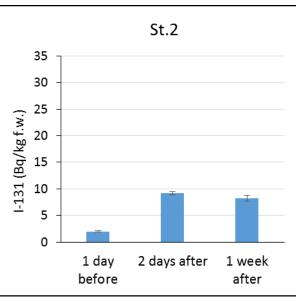
Figure 4. Location of sampling stations in 2016 in relation to discharge point for effluent from the University Hospital of North Norway and the Breivika WTP.

Activity concentrations of I-131 were observed at all three stations both before and after the patient was treated (Fig.5), but with higher activity concentrations observed after the patient was treated. The highest activity concentration of I-131 (31.5  $\pm$ 1.3 Bq/kg f.w.) was observed at St.3, two days after the patient was treated. St.3 is closest to the discharge point for effluent from Breivika WTP and so would be expected to be exposed to higher activity concentrations in the receiving seawater as a result of any discharges than

compared to St.1 and St.2. However, it is not known at which point in the tidal cycle that the discharges of I-131 occurred.

Further sampling of *F. vesiculosus* was carried out in February 2017 to determine the uptake in different parts of the seaweed and to examine the effect of drying samples before analysis. A large sample of *F. vesiculosus* were collected at St.3, three days after a patient was treated with 6 GBq of I-131. The collected seaweed was separated into sub-samples of blades and stipes with one part of each sub-sample analysed fresh weight whilst the other part was dried at 60 °C over night before analysis.





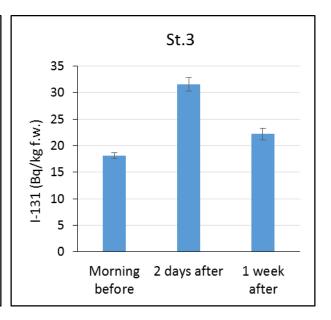


Figure 5. Activity concentrations of I-131 (Bq/kg f.w.) in *F. vesiculosus* at 3 sampling stations in the area around the discharge point for effluent from Breivika WTP at stated times before and after a patient was admitted for I-131 radiotherapy (4 GBq of I-131) at UNN. The last previous patient admitted for radiotherapy with I-131 at UNN was 20 days earlier.

The activity concentrations of I-131 in blades of *F. vesiculosus* was higher than in stipes by a factor of 2 to 3 (Table 2). The impact of drying would suggest that there is some loss of I-131 from the blades but not from the stipes. This is most likely due to loss of cytosol from the blades to the drying tray during the drying process. The fresh weight activity concentration of I-131 in the samples collected in February 2017 were almost a factor of 10 higher than was observed in August 2016. This difference cannot be explained alone by the fact that the patient in August 2016 was treated with 4 GBq compared to the 6 GBq received by the patient in February 2017. Instead it is likely that the tidal cycle and current direction are the controlling factors on the exposure of seaweed to any I-131 discharges. With regard to the tidal cycle, if any discharge occurs at low tide, any seaweed in the intertidal zone at St.3 would not be exposed until later in the tidal cycle and then at possibly lower activity concentrations due to effects of dilution by current mixing. However, where discharges of I-131 occur at high tide, seaweed in the intertidal zone at St.3 would then be exposed to maximal activity concentrations, depending on the current direction at that point in time.

Table 2. Activity concentrations of I-131 in parts of F. vesiculosus from St.3, analysed fresh or after drying.

d.w./f.w.	l-131 (Bq/kg d.w.)	±	I-131 (Bq/kg f.w.)	±
-	-	-	292	15
-	-	-	114	7
0.23	1040	52	242	12
0.35	360	18	126	6
	0.23	0.23 1040		- - - 292   - - - 114   0.23 1040 52 242

Further sampling of *F. vesiculosus* was carried out in March and April 2017 at an additional nine sampling stations to determine the geographical extent over which I-131 discharged from the Breivika WTP could be detected in seaweed (Fig.6).

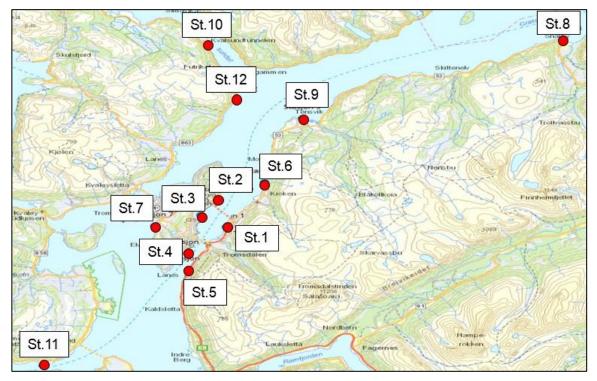


Figure 6. Locations of stations 1 to 12 for sampling of F. vesiculosus in March and April 2017.

Samples of *F. vesiculous* were collected over a period of four weeks, beginning one day before a patient was treated with 6 GBq of I-131 at UNN. A second patient received a further 6 GBq a week after the first patient. Prior to the start of sampling the last previous patient admitted for radiotherapy with I-131 at UNN was four weeks earlier. Activity concentrations of I-131 above detection limits were only observed at St.3 (14.2 ±1.4 Bq/kg f.w.) in the initial round of sampling.

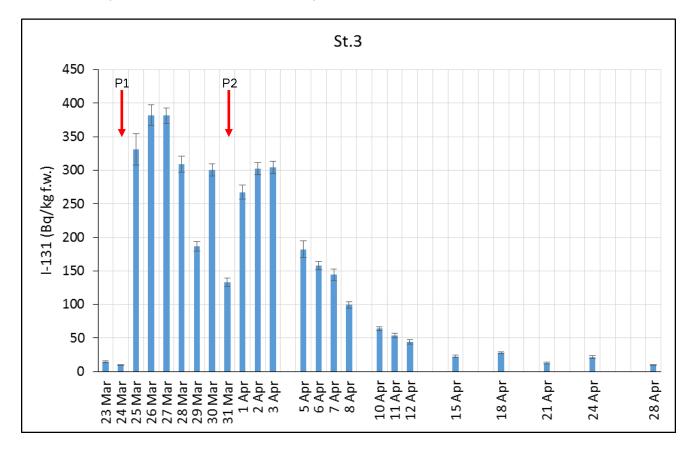


Figure 7. Activity concentrations of I-131 (Bq/kg f.w.) in F. vesiculosus at St.3 in March and April 2017. Individual patients (red arrows) were treated with 6 GBq of I-131 on the  $24^{th}$  and  $31^{st}$  of March.

Activity concentrations of I-131 in F. vesiculosus at St.3 increased sharply from 9.6  $\pm$ 0.5 on the morning of the 24th of March to 331  $\pm$ 23 Bq/kg f.w. 30 hours later after the first patient was treated with 6 GBq of I-131, peaking at 381  $\pm$ 15 Bq/kg f.w. a further 33 hours later (Fig.7). Activity concentrations of I-131 in F. vesiculosus at St.3 then decreased over the next five days to 187  $\pm$ 7 Bq/kg f.w. before the second patient was treated with a further 6 GBq of I-131. A higher than expected activity concentration of I-131 was observed on the 30th of March which might reflect variations in exposure of different patches of F. vesiculosus at the same sampling station. Following the treatment of the second patient, activity concentrations of I-131 in F. vesiculosus at St.3 increased again to 304  $\pm$ 9 Bq/kg f.w. on the 3rd of April. Activity concentrations of I-131 in F. vesiculosus at St.3 then decreased over the next 25 days to 9.5  $\pm$ 0.5 Bq/kg f.w., the same value as was observed prior to the treatment of the first patient on the 24th of March. It is interesting to note that increase in activity concentrations of I-131 in F. vesiculosus at St.3 was less following the treatment of the second patient compared to the first patient, although both were treated with 6 GBq of I-131. This is probably a reflection of the combination of different excretion rates of I-131-from the two patients and the times at which discharges occurred with regard to the status of the tidal cycle and the direction of current flow in Tromsøysundet.

Following the treatment of the first patient on the  $24^{th}$  of March, samples of *F. vesiculosus* were collected at Stations 1 to 7 on five different days over a four week period (Fig.8). Activity concentrations of I-131 at St.2 ranged from 174  $\pm$ 7 Bq/kg f.w. on the 27th March to  $3.1 \pm 0.3$  Bq/kg f.w. on the  $18^{th}$  of April. Any observations of I-131 activity concentrations in *F. vesiculosus* from Stations 1 and 4 to 7 were below 10 Bq/kg f.w.. Activity concentrations of I-131 above detection limits were first detected in the samples collected at Stations 2 and 4 on the on  $27^{th}$  of March, at Stations 1 and 6 on the  $30^{th}$  of March and at Stations 5 and 7 on the  $7^{th}$  of April. Samples of *F. vesiculosus* collected at Stations 8 to 12, 5 and 20 days after the second patient was treated, did not show any activity concentrations above detection limits (<0.6 to <2.7 Bq/kg f.w.).

The trend in observations would suggest that the discharges of I-131 are transported along the western side of Tromsøysundet, before dispersal across to the eastern side. Coupled with the previous observations of activity concentrations of I-131 in *F. vesiculosus* at the Grøtsund Port Facility (St.9), the main direction of transport of I-131 discharges would appear to be northwards. That activity concentrations of I-131 were detected at St.7 on the opposite side of Tromsøya would indicate that discharges of I-131 are transported from Tromsøysundet through the northern and/or southern approaches of Sandnessundet.

For comparison purposes, three different species of edible brown seaweed were collected at St.3 on the 28<sup>th</sup> of March, four days after the first patient was treated with 6 GBq of I-131. Compared to the sample of *F. vesiculosus* sampled at the same time, activity concentrations of I-131 were higher in *Laminaria* saccharina but lower in *F. evanescens* and *Ascophyllum nodosum* (Table 3). As *L. saccharina* is a sublittoral species it would be expected to be exposed to discharges of I-131 regardless of the tidal cycle, which may account for the observed higher activity concentration of I-131 compared to *F. vesiculosus*. *F. evanescens* and *A. nodosum* are both intertidal species and both were sampled within the same horizontal band as *F. vesiculosus*.

Table 3. Activity concentrations of I-131 (Bq/kg f.w.) in brown seaweed species at St.3 sampled on the 28<sup>th</sup> of March 2017.

Seaweed	I-131 (Bq/kg f.w.)	±
Fucus vesiculosis	309	12
Ascophyllum nodosum	151	8
Laminaria saccharina	383	12
Fucus evanescens	229	9

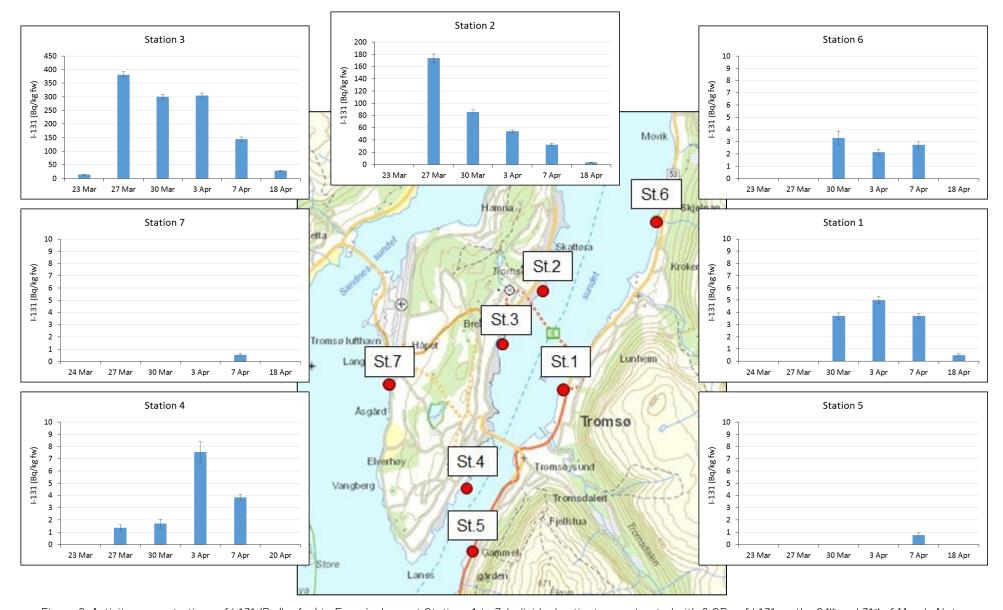


Figure 8. Activity concentrations of I-131 (Bq/kg f.w.) in *F. vesiculosus* at Stations 1 to 7. Individual patients were treated with 6 GBq of I-131 on the 24<sup>th</sup> and 31<sup>st</sup> of March. Note different scales on vertical axes.

Further sampling of F. vesiculosus at St.3 was carried out in August 2017 in association with a patient treated with 4 GBq of I-131 (Fig.9). Prior to the start of sampling the last previous patient admitted for radiotherapy with I-131 at UNN was five weeks earlier. The day before the patient was treated, the activity concentration of I-131 in F. vesiculosus at St.3 was 7.7  $\pm$ 0.5 Bq/kg f.w.. Three days after the patient was treated, the activity concentration in F. vesiculosus at St.3 was 157  $\pm$ 3 Bq/kg f.w. decreasing 4 days later to 71  $\pm$ 4 Bq/kg f.w..

A single sample of F. vesiculosus was collected at St.3 in September 2017, three days after a patient was treated with 6 GBq of I-131. Prior to the treatment of this patient, the last previous patient admitted for radiotherapy with I-131 (3.7 GBq) at UNN was just six days earlier. The activity concentration of I-131 in this sample of F. vesiculosus was 732  $\pm$ 29 Bq/kg f.w..

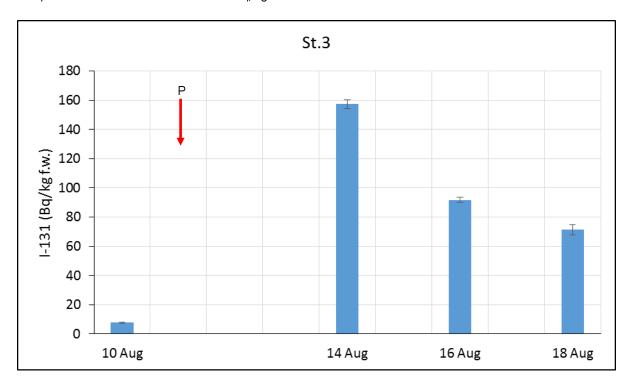


Figure 9. Activity concentrations of I-131 (Bq/kg f.w.) in F. vesiculosus at St.3 in August 2017. A patient (red arrow) was treated with 4 GBq of I-131 on the  $11^{th}$  of August.

Further sampling of F. vesiculosus at St.3 was carried out in October 2017 in association with a patient treated with 6 GBq of I-131 (Fig.10). Three days before the start of sampling another patient had been treated with 6 GBq of I-131 at UNN and a further week before that, another patient had also been treated with 6 GBq of I-131. The activity concentration of I-131 in F. vesiculosus on the morning of the  $16^{th}$  of October was 446  $\pm 18$  Bq/kg f.w., reflecting the exposure of seaweed at this station from discharges of I-131 associated with the two previous patients. Following the treatment of a further patient on the  $17^{th}$  of October, activity concentrations of I-131 in F. vesiculosus at St.3 then peaked at  $682 \pm 27$  Bq/kg f.w. on the morning of the  $19^{th}$  of October before decreasing to  $337 \pm 13$  Bq/kg f.w. four days later.

The observations from September and in particular October 2017 illustrate the potential for additive effects of consecutive discharges of I-131 on activity concentrations of I-131 in *F. vesiculosus* at St.3, when successive patients are treated for radiotherapy with I-131. In the case, of October 2017 three patients were each treated with 6 GBq over a period of only nine days. The activity concentrations of I-131 in seaweed observed in this study were typically higher than reported values from similar studies that examined the uptake by seaweed of I-131 discharged from hospitals. Activity concentrations of I-131 in *F. vesiculosus* of up to 61 Bg/kg f.w have been reported in the 1990s at one coastal location in Ireland that

was influenced by discharges from a hospital (e.g. Pollard et al., 1996). More recently, Sundell-Bergman et al. (2009) reported activity concentrations of I-131 from hospital discharges in two samples of *F. vesiculosus* of 13 ±2 and 26 ±3 Bq/kg f.w. in Finland. Similarly, Carolan et al. (2011) reported activity concentrations of I-131 of 0.5 to 38 Bq/kg f.w. in a green (*Ulva* spp.) seaweed and 20 to 70 Bq/kg f.w. in a brown (*Ecklonia radiata*) seaweed as a result of discharges from hospitals and a research centre in Sydney, Australia. Meanwhile, Kelly et al. (2014) reported typical activity concentrations of I-131 in the range of 2 to 20 Bq/kg f.w. (assuming a d.w./f.w. ratio of 0.2) for intertidal seaweeds (*A. nodosum*, *Saccharina latissimi* and *F. garneri*) resulting from hospital discharges in Halifax, Canada, but with a peak value of 3000 Bq/kg f.w. No other radioisotopes in use at UNN were observed at levels above detection limits in any *F. vesiculosus* sample collected in this study.

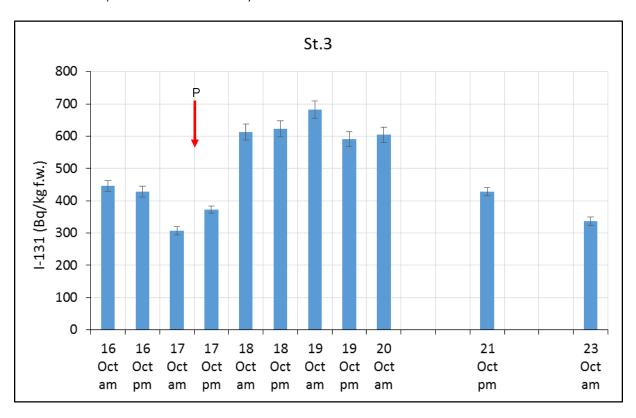


Figure 10. Activity concentrations of I-131 (Bq/kg f.w.) in F. vesiculosus at St.3 in October 2017. A patient (red arrow) was treated with 6 GBq of I-131 on the  $17^{th}$  of October. Additional patients had been treated with 6 GBq of I-131 on the  $6^{th}$  and  $13^{th}$  of October.

#### 2.3 Effective and biological half lifes of I-131 in F. vesiculosus

Effective half lifes ( $T_{eff}$ ) for I-131 in F. vesiculosus can be derived from data from March and April for St.3 and St.2 using equation 1.

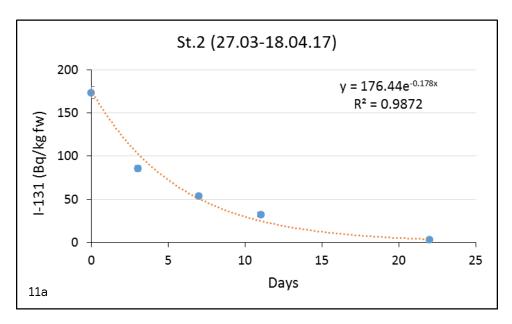
Equation 1:

$$T_{eff} = -ln(2)/a$$

Where a is the exponent of the best fit exponential equations in Figures 11a,b.  $T_{\rm eff}$  for St.2 and St.3 on the stated dates were 3.9 and 5.2 days, respectively. Biological half lifes ( $T_{\rm bio}$ ) for I-131 can be derived from these  $T_{\rm eff}$  values using equation 2.

$$1/T_{eff} = 1/T_{phys} + 1/T_{bio}$$

Where  $T_{phys}$  is the physical half-life of I-131 of 8.0197 days.  $T_{bio}$  for St.2 and St.3 on the stated dates were 7.6 and 14.6 days, respectively.



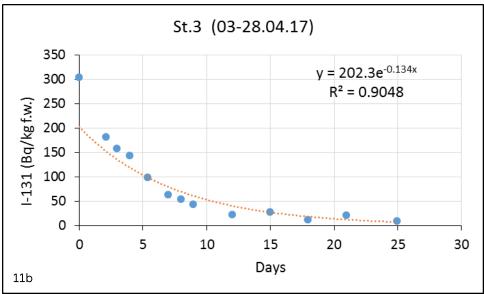
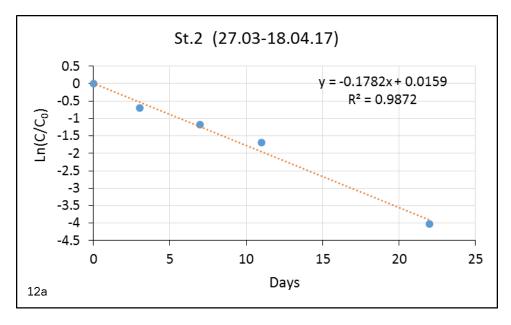


Figure 11a and b. Activity concentrations of I-131 in F. vesiculosus at St.2 and St.3 on stated dates for the derivation of  $T_{\rm eff}$ .

For both St.2 and St.3, it might be expected that the seaweed at these stations would be continually exposed to decreasing activity concentrations of I-131 in seawater as the discharges are dispersed and diluted. This may be particularly apparent at St.3 which is closest to the discharge point. To determine if this was the case the logarithm of I-131 activity concentrations in *F. vesiculosus* relative to the initial concentrations were plotted for each case (Fig.12a,b)



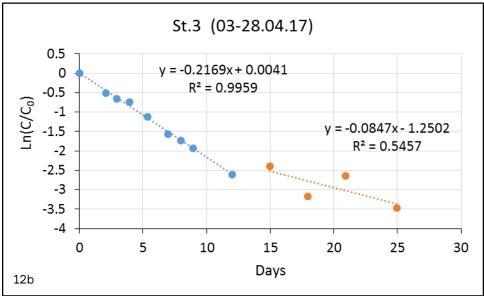


Figure 12a and b. Logarithm of I-131 activity concentrations in F. vesiculosus relative to the initial concentrations at St.2 and St.3 on stated dates for the derivation of  $T_{\rm eff}$ .

For St.2, there was no difference to the derivation of  $T_{\rm eff}$  and therefore  $T_{\rm biol}$  between the approach used in Figure 11a and 12a. For St.3, it was clear that there were 2 phases to the overall period (Fig. 12b), with the first phase yielding an improved  $R^2$  value of 0.9959 compared to a  $R^2$  value of 0.9048 when considering the entire data period as in Figure 11b. The second phase yielded a lower  $R^2$  value of just 0.5457, which reflects the fluctuation in activity concentrations that were observed during this period. These fluctuating activity concentrations may have resulted from further exposure of F.  $V_{\rm esiculosus}$  at St.3 to I-131 in seawater, or they may simply reflect natural variations as a result of sampling. Using just the data from the first phase, values of  $T_{\rm eff}$  and  $T_{\rm bio}$  of 3.2 and 5.3 days can be derived, which are in better agreement with the values of  $T_{\rm eff}$  and  $T_{\rm bio}$  derived from data from St.2. The values derived in this study can be compared with a previously reported  $V_{\rm bio}$  for  $V_{\rm eff}$  and  $V_{\rm bio}$  for  $V_{\rm eff}$  and 10.3 days (Druehl et al., 1988; Vives i Batlle et al., 2008) and of 5 to 21 days for 2 species of the closely related  $V_{\rm eff}$  seaweed (Hevano et al., 1983; Guimaraes & Penna-Franca, 1985).

#### 2.4 Dose assessment for *F. vesiculosus* and human pathways

It is clear that activity concentrations of I-131 in F. vesiculosus and in the surrounding water fluctuate in relation to the frequency to which patients are admitted for radiotherapy treatment with I-131 at UNN. The maximum activity concentration of I-131 observed in F. vesiculosus at any time at St.3 was 732 ±29 Bq/kg f.w., but on other occasions, activity concentrations were lower than 10 Bq/kg f.w.. No data is available for activity concentrations of I-131 in seawater for St.3, however it is possible to estimate an initial seawater activity concentration following any discharge in the first 24 hours after a patient has been treated with I-131. Using the recommended macroalgae biological concentration factor for iodine of 1 x 10<sup>4</sup> (IAEA, 2004) and the increase in activity concentrations between the 24th (9.6 Bq/kg f.w.) and 25th (331 Bq/kg f.w.) of March following the treatment of a patient with 6 GBq of I-131, a back calculated seawater I-131 activity concentration of 0.032 Bq/l can be derived. No exposure from sediments is assumed as F. vesiculosus grows on rock surfaces. From the maximum observed I-131 activity concentration in F. vesiculosus at St.3 and the back calculated seawater l-131 activity concentration, a total dose rate of  $8.14 \times 10^{-2} \,\mu\text{Gy/hr}$  can be determined for F. vesiculosus at St.3 using the Erica tool (Brown et al., 2008). This total dose rate is dominated by the internal dose rate from bioacummulated I-131 and is three orders of magnitude lower than the ERICA dose rate screening value for marine biota of 10 µGy/hr (Garnier-Laplace et al., 2008) below which no effects would be predicted to occur.

In terms of dose assessments to humans, it is highly unlikely that anyone would swim in the sea or occupy the area around St.3 for any length of time as this location is in an industrial area and is there is no defined public access. UNN have already estimated an occupancy dose rate for the harbour area in Breivika of 170 μSv/a. Observed I-131 activity concentrations in F. vesiculosus at St.3 regularly exceeded 100 Bg/kg which is the guideline limit for I-131 in food stuffs following a nuclear or radiological emergency (FAO/WHO, 2006). However, although F. vesiculosus is edible the daily amount that can be consumed safely is low due to the potential for hyperthyroidism through excess iodine ingestion. Typically suggested daily dosages for commercially available dried F. vesiculosus preparations are up to 2 g per day. As observed, activity concentrations of I-131 in F. vesiculosus at St.3 can vary by 2 orders of magnitude and these would decay further by T<sub>phys</sub> in any F. vesiculosus harvested for human consumption. In terms of exposure via the ingestion pathway it is unlikely that anyone would harvest F. vesiculosus from St.3 for personal consumption. However, by taking an extremely conservative approach and assuming a constant I-131 activity concentration of 732 Bq/kg f.w., i.e. no loss by physical decay, a daily dosage of 2 g of dried F. vesiculosus and the adult ingestion dose coefficients for I-131 of 0.022 μSv/Bq (IAEA, 1996), then a hypothetical maximum annual ingestion dose rate of 47 µSv/a can be derived. Despite the extremely conservative approach, this derived annual ingestion dose rate is still 2 orders of magnitude lower than the individual dose limit for public exposure of 1 mSv/a.

#### 2.5 Sampling and analysis of sewage samples

Samples of influent, effluent and solid waste were sampled at the Breivika WTP. Samples of influent and effluent were collected immediately before and after filtration at the Breivika WTP as composite samples using in situ automatic samplers that sampled 40 ml every 8 minutes. The time taken for incoming influent to travel from the influent sampling point, via the filtration units to the effluent sampling point is estimated to be of the order of 1 minute (Pers. comm. Tord Bentzen). Composite samples were made over either 8, 16 or 24 hours. Solid waste was sampled directly from the filtrate collector of the filtration units at the Breivika WTP. However, it is not possible to know when the samples of solid waste were filtered out from influent, but it is assumed they represent an integrated time period from with the previous period of the influent and effluent composite samples. The filtration units and filtrate collector at the Breivika WTP are cleaned at least 3 times a week on Mondays, Wednesdays and Fridays. All sewage samples were analysed immediately upon return to the laboratory. Influent and effluent samples were filtered through glass microfibre filters (pore size 1.6 µm) before analysis with results reported as Bq/l. Solid waste samples were not dried before analysis and results are given as Bq/kg f.w.. After analysis, solid waste samples were dried at 105 °C overnight to determine the water content. Dry weight/fresh weight ratios for solid waste samples typically ranged from 0.15 to 0.27. All samples were packed into plastic counting geometries and were analysed for at least 1 hour on HPGe detectors. Results for influent and effluent were decay corrected to the midpoint of the composite collection period, while results for solid waste were decay corrected to the sampling time and date. All results are given with associated uncertainties to 1 sigma.

#### 2.6 Results for I-131 in sewage samples

Initial screening of sewage samples were conducted on two occasions in conjunction when patients were treated with I-131 at UNN. 24 hr composite samples of influent and effluent were collected starting at 13:00 pm on the day the patients were treated. Patients normally begin I-131 radiotherapy between 10:00 and 11:00 am (Pers. comm. Anders Eide Lund). These composite samples were collected and analysed the following Monday. Samples of solid waste were collected on the Monday morning on each occasion, before scheduled cleaning of the filtrate collector.

Activity concentrations of I-131 in influent and effluent in each sample pair were similar, with higher activity concentrations in influent and effluent samples when the patient was treated with 6 GBq compared to treatment with 3.7 GBq (Table 4). That similar activity concentrations were observed in the influent and effluent samples would suggest that the I-131 remains in solution as it passes through filtration process at Breivika WTP. Activity concentrations of I-131 in solid waste were somewhat lower in the sample taken after a patient received 6 GBq compared to the sample taken after a patient received 3.7 GBq (Table 4). This may appear counterintuitive, but the solid waste sample taken after a patient received 6 GBq contained half the amount of water (d.w./f.w. of 0.42) as compared to the other solid waste sample (d.w./f.w. of 0.20). If I-131 remains in solution, then the amount of I-131 in the solid waste may be controlled by the amount of influent water present in the waste when it is collected.

From such composite sampling it is not possible to determine how many I-131 pulses pass through the Breivika WTP in any given sampling period, nor how long it takes for an I-131 pulse to pass the respective influent and effluent sampling points, nor whether the activity concentrations in the pulses are heterogeneous. Therefore, it is possible that pulses are either missed completely if any pulse length is less than the sampling interval of 8 minutes (or if any pulse occurs before sampling begins) and or that activity concentrations of each pulse may be underrepresented in the composite samples.

Table 4. Activity concentrations of I-131 in composite influent and effluent samples (Bq/I) and solid waste (Bq/kg f.w.) collected at Breivika WTP.

Date & dosage of I-131	Sampling time/Sample	I-131 (Bq/I)	±
18/08/17 - 3.7 GBq	13:00 18/08/17 to 13:00 19/08/17	(	
· · ·	Influent	284	11
	Effluent	268	8
	08:45 21/08/17		
	Solid waste	151	6
01/09/17 - 6 GBq			
	13:00 01/09/17 to 13:00 02/09/17		
	Influent	460	18
	Effluent	474	14
	08:30 04/09/17		
	Solid waste	130	5

However, assuming that all pulses were sampled and that activity concentrations of each pulse were homogenous, it is possible to derive the total activity excreted based on the total volume of influent that passed through the WTP during the sampling period. For this purpose, the mean activity concentration of each influent and effluent composite sample pair was used to reduce potential sampling uncertainties. Estimated percentages of I-131 excreted into the sewage system 24 hr after patients received 3.7 GBq and 6 GBq were 79.8% and 60.0%, respectively (Table 5). According to Mettler and Guiberteau (2012), 35% to 75% of iodine is excreted from the body in the first 24 hours, predominately in urine. In addition, Demir et al. (2013) reported that on average 66% of I-131 administered is excreted in urine after 24 hours, with 87% excreted after 48 hours. However, it is expected that amount of I-131 excreted within 24 or 48 hours will vary between patients depending on the degree of initial uptake and any variations in biological half lifes for absorbed I-131. To illustrate this issue, UNN has previously observed that the excretion of I-131 after 48 hours for 8 patients varied between 62% and 92% (Pers. comm. Anders Eide Lund).

Table 5. Estimation of amount of I-131 excreted in 24 hour sampling periods following radiotherapy treatment of patients with I-131.

Dosage (GBq)	Mean I-131 in composite samples (Bq/I)	Volume of influent in 24 hr (m³)	Activity excreted in 24 hr (GBq)	Percentage excreted
3.7	276	10701.4	3.0	79.8%
6	467	7712.2	3.6	60.0%

Based on the screening results above, more in depth monitoring of I-131 and other isotopes in use by UNN in sewage samples at Breivika WTP was then carried out. In the period from the 13<sup>th</sup> to the 20<sup>th</sup> of October

2017, two patients were each treated with 6 GBq of I-131 and a number of other patients were given varying activities of Tc-99m, I-123 and F-18 for diagnostic radiography purposes (Table 6).

Following an initial composite sampling period of 24 hr on Friday the 13<sup>th</sup> of October that commenced at 13:00 pm, 8 hr and 16 hr composite samples were collected between 8:00 am and 16:00 pm (day) and 16:00 pm and 8:00 am (night), respectively, from Monday to Friday of the following week. In addition, solid waste samples were collected at 8:00 am and 16:00 pm each day. Unfortunately, the Breivika WTP was offline on the 17th of October, the same day that a second patient was treated with 6 GBq of I-131, so no sewage samples were available for this 24 hr period.

Table 6. Number of patients and total activities of I-131, Tc-99m, I-123 and F-18 used between the  $13^{th}$  of October and  $20^{th}$  of October 2017 at UNN.

Day	I-	131	Tc-	99m	I-1	L23	F-:	
	n	GBq	N	GBq	n	GBq	n	GBq
Fri	1	6	NA		NA			NA
Mon			10	3.6				
Tue	1	6	2	0.2			8	2.0
Wed			3	1.5				
Thu			4	1.3	2	0.37	8	2.0
Fri	·		2	1.5				

For the initial 24hr composite sampling, data is only available for influent as the effluent automatic sampler failed to function. All subsequent influent and effluent composite sample pairs showed similar I-131 activity concentrations, as had been previously observed, supporting the suggestion that I-131 remains in solution as it passes through the WTP.

The activity concentration of I-131 in the influent 24 hr composite sample was 743 ±22 Bq/l, a factor of 1.6 higher than was observed in the previous 24 hr composite sample collected after a patient received 6 GBq. This patient was discharged the following Monday, but I-131 was still detected in the influent and effluent composite samples that were collected during the day (Fig.13). The subsequent influent and effluent composite samples collected on the Monday night showed no I-131 activity concentrations above detection limits. Assuming the same conditions as previously, the estimated percentage of I-131 excreted into the sewage system from this patient in the first 24 hr was 101.7%, while the percentage of I-131 excreted during the day on Monday was 1.4% (Table 7). No information is available concerning the particular treatment of this patient, but these observations could be explained by a low uptake of I-131 by the patient (e.g. if the patient vomited before the I-131 capsule was fully dissolved). Alternatively, this finding may demonstrate the limitations of this approach to estimate the percentage of I-131 excreted. The solid waste sample collected on Monday morning had an I-131 activity concentration of 1329 ±27 Bq/kg f.w., compared to 298 ±9 Bq/kg f.w. for the sample collected 8 hr later (Fig. 14).

The activity concentration of I-131 in the influent and effluent 8 hr composite samples collected approximately 24 hr after the second patient received 6 GBq were 217  $\pm$ 4 and 229  $\pm$ 7 Bq/I, respectively.

Activity concentrations of I-131 in subsequent composite samples collected over the next days decreased, with values of  $1.1 \pm 0.1$  and  $0.9 \pm 0.1$  Bq/I observed for the composite influent and effluent samples collected on the Friday night after the patient had been discharged from UNN (Fig.13). The total estimated percentage of I-131 excreted into the sewage system from this patient between Wednesday to Friday was 28.3% (Table 7), which implies that up to 69.8% of the 6 GBq that was given to the second patient was excreted in the first 24 hr. This value is comparable to the previous 24 hr excretion estimates of 60.0% and 79.8% in this study and reported excretion rates of I-131 (Mettler and Guiberteau, 2012; Demir et al., 2013).

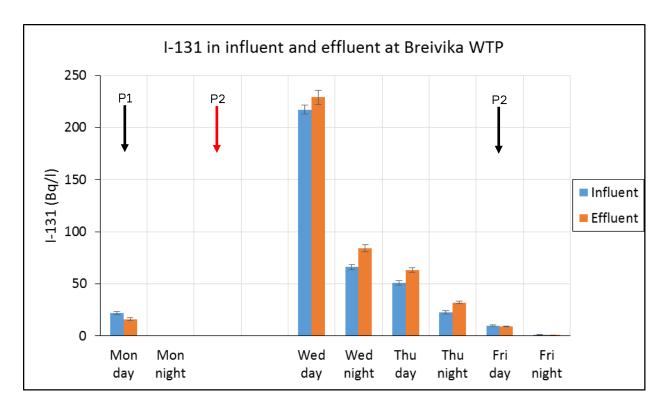


Figure 13. Activity concentrations in I-131 (Bq/I) in composite influent and effluent sampled at Breivika WTP during the day (8 hr) and at night (16 hr) over the course of 1 week. No sampling was possible on the Tuesday as the Breivika WTP was offline. Black arrows indicate when patients were discharged from UNN after radiotherapy treatment and the red arrow indicates the day a patient was treated with 6 GBq of I-131.

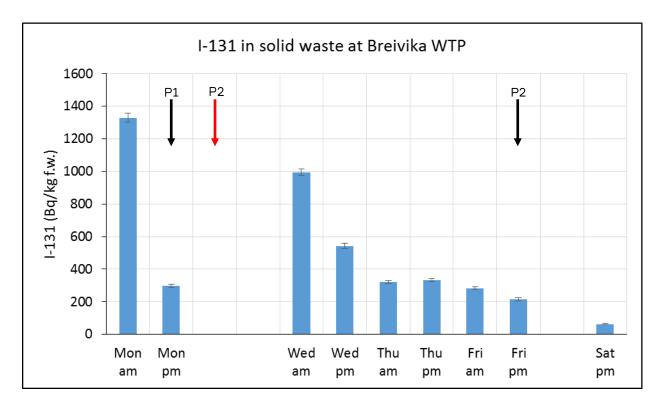


Figure 14. Activity concentrations of I-131 (Bq/kg f.w.) in solid waste sampled at Breivika WTP in the morning and afternoon over the course of 1 week. No sampling was possible on the Tuesday as the Breivika WTP was offline. Black arrows indicate when patients were discharged from UNN after radiotherapy treatment and the red arrow indicates the day a patient was treated with 6 GBq of I-131.

Table 7. Estimation of amount of I-131 excreted in composite sampling periods following radiotherapy treatment of 2 patients with 6 GBq of I-131.

Sampling		Mean I-131 in composite samples (Bq/I)	Volume of influent (m³)	Activity excreted (GBq)	Percentage excreted	
Period			, ,	ν μ		
Fri/Sat	24	743°	8221	6.1	101.7%	
Mon day	8	19	3898	0.07	1.2%	
Mon night	16	<2.5	8278	-	-	
Wed day	8	223	3440	0.77	12.8%	
Wed night	16	71	8123	0.58	9.6%	
Thu day	8	57	4266	0.24	4.1%	
Thu night	16	27	6829	0.19	3.1%	
Fri day	8	9.6	3340	0.03	0.5%	
Fri night	16	1.0	6170	0.01	0.1%	

a - based on activity concentration from influent composite sample only, due to the failure of the effluent automatic sampler.

Activity concentrations of I-131 in solid waste samples collected over the same period after the second patient was treated with 6 GBq of I-131 showed a similar trend as for influent and effluent composite samples (Fig. 14). Activity concentrations of I-131 in the solid waste sample collected on Wednesday morning had an I-131 activity concentration of  $994 \pm 20$  Bq/kg f.w., decreasing to  $215 \pm 11$  Bq/kg f.w. for the sample collected on Friday afternoon. It is interesting to note that after the second patient was discharged on the Friday, and assuming that scheduled routine cleaning of the filtrate collector occurred on the same day, that activity concentrations of I-131 (62  $\pm 4$  Bq/kg f.w.) were still detectable in a solid waste sample collected on Saturday afternoon (Fig. 14). This would suggest that the residence time for solid waste in the filtrate collector can be at least 24 hr.

#### 2.7 Results for Tc-99m in sewage samples

Tc-99m is used at UNN for diagnostic radiography on a daily basis from Monday to Fridays but only between 07:30 am and 15:00 pm, with the total amount used on any day dependent on the number of patients and the activities of Tc-99m given to each patient. No activity concentrations of Tc-99m above detection limits (<1.4 to <5.6) were observed in any of the initial screening samples of influent, effluent or solid waste. If any patients had received Tc-99m on the days (Friday) when 24 hr composite sampling began, any Tc-99m collected in either the influent or effluent samples would have decayed entirely (half-life of 6 hours) by the time the samples were analysed 3 days later (Monday).

When more in depth monitoring was carried out, activity concentrations of Tc-99m were observed in all influent and effluent composite samples collected during the day, while values were typically below

detection limits in the composite samples collected during the night (Fig.15). This pattern reflects the use of Tc-99m at UNN between 07:30 am and 15:30 pm only. In composite samples collected during the day, Tc-99m activity concentrations were similar for each influent and effluent pair, with a range of 37  $\pm$ 2 to 47  $\pm$ 3 Bq/l. As was the case for I-131, this would suggest that Tc-99m remains in solution as discharges pass through the Breivika WTP.

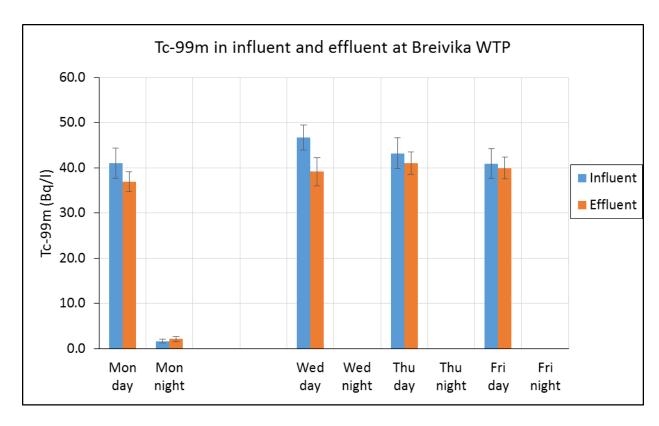


Figure 15. Activity concentrations in Tc-99m (Bq/I) in composite influent and effluent sampled at Breivika WTP during the day (8 hr) and at night (16 hr) over the course of 1 week. No sampling was possible on the Tuesday as the Breivika WTP was offline.

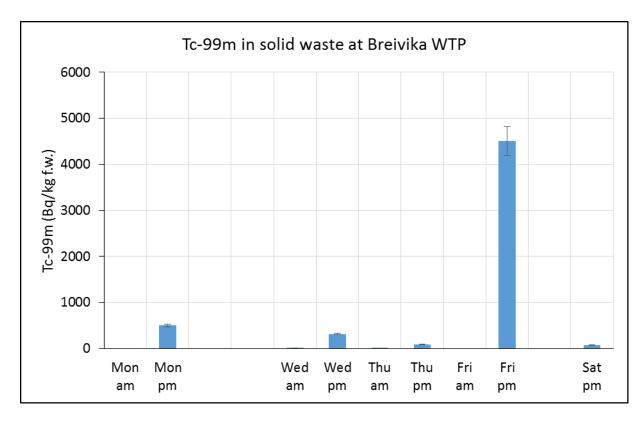


Figure 16. Activity concentrations of Tc-99m (Bq/kg f.w.) in solid waste sampled at Breivika WTP in the morning and afternoon over the course of 1 week. No sampling was possible on the Tuesday as the Breivika WTP was offline.

Table 8. Estimation of amount of Tc-99m excreted at UNN during the day.

Sampling		Mean Tc-99m in	Volume of influent	Activity excreted	Percentage
Period	Time (hr)	composite samples (Bq/I)	(m³)	(GBq)	excreted <sup>1</sup>
Mon	8	39	3898	0.15	4.3%
Wed	8	43	3440	0.15	9.6%
Thu	8	42	4266	0.18	13.8%
Fri	8	40	3340	0.13	9.0%

<sup>1 -</sup> as a percentage of the total activity used on these days as stated in Table 6.

Using the same approach as for I-131 and in comparison with the total activity used each day, the percentage of Tc-99m excreted to Breivika WTP ranged between 4.3% and 13.8% (Table 8). These values are somewhat lower than the recommended excretion factor of 40% for Tc-99m for its use in diagnostic radiography (IPEM, 2018). However, as patients who receive Tc-99m are not isolated, it is not expected that all excretion of Tc-99m will occur at UNN or to the same sewage system.

Higher activity concentrations of Tc-99m were observed in solid waste samples collected in the afternoon (89  $\pm 4$  to 4507  $\pm 315$  Bq/kg f.w.) compared to the morning (<2.9 to 8.2  $\pm 1.1$  Bq/kg f.w.) for the same day (Fig.16). This pattern again reflects the use of Tc-99m between 07:30 am and 15:30 pm only. In addition, the observation of low activity concentrations above the detection limit in solid waste samples collected in the morning might further support a relatively long residence time (>16 hr) for solid waste in the filtrate collector. i.e. if Tc-99m observed in samples collected in the morning is derived from discharges that occurred the day before. Alternatively, the presence of Tc-99m in solid waste samples collected in the morning may reflect excretions by patients who were required to stay at UNN or who have returned to a household within the same sewage system area as UNN.

#### 2.8 Results for other radioisotopes in sewage samples

During the week of more in depth monitoring, a number of patients at UNN received F-18 (Tuesday and Thursday) and I-123 (Thursday). F-18 and I-123 are used for diagnostic radiography and have relatively short half lifes of 110 minutes and 13.2 hours, respectively. As previously mentioned, no sampling was possible at the Breivika WTP on the Tuesday during this week. However, on the Thursday, activity concentrations of I-123 were observed in composite samples of influent (8.4  $\pm$ 1.1 Bq/l) and effluent (5.8  $\pm$ 0.9 Bq/l) collected during the day. Furthermore, I-123 was detected in the solid waste sample collected on the Thursday afternoon (5.9  $\pm$ 1.0 Bq/kg f.w.). Using the same approach as for I-131 and the information in Tables 6 and 8, the percentage of I-123 excreted to Breivika WTP can be estimated at 8.2%. The recommended excretion factor for the form of I-123 given to these patients (MIBG) is 30% (IPEM, 2018) and it is expected that I-123 will follow the same excretion pathways as for I-131. However, as patients who receive I-123 are not isolated, it is not expected that all excretion of I-123 will occur at UNN or to the same sewage system. No activity concentrations of I-123 were detected above detection limits in the composite influent (<1.7 Bq/l) and effluent (<1.8 Bq/l) samples collected the following night, nor in the solid waste sample (<3.6 Bq/kg f.w.) collected on the Friday morning.

F-18 is a beta emitter that decays by positron emission 97% of the time and electron capture 3% of the time. Interaction of the emitted positrons with electrons generates annihilation gamma emissions at 511

keV. A marked increase in the expected counts per minute for the typical annihilation peak at 511 keV for such samples was observed for the solid waste sample collected on the Thursday afternoon (125.4 cpm). When this sample was reanalysed 4 days later, the counts per minute for the 511 keV annihilation peak had decreased to 1.15 cpm, a value similar to annihilation peaks for all other solid waste samples collected at Breivika WTP. If one assumes that the only contribution to the 511 keV peak above background is due to F-18, a F-18 activity concentration of 772 ±23 Bq/kg f.w. can be obtained for this solid waste sample. No activity concentrations of F-18 above detection levels were observed in the influent (<24 Bq/l) and effluent (<30 Bq/l) composite samples collected the same day.

# 3 Consideration of possible exposures and doses to workers at Breivika WTP

As stated previously, UNN has carried out an assessment of doses to sewage workers at the Breivika WTP. In this assessment, a sewage worker carried a dosimeter for 16 working days and recorded an average dose of 0.033 µSv/hr, equivalent to 67 µSv/a. However, it is not known what work practices the sewage worker carried out or whether any these work practices coincided with any patients receiving I-131 or any other radioisotopes during the 16 day period. Potential exposures of workers at Breivika WTP to discharges of radioisotopes of UNN can occur from being present in the filtration room during normal operation of the WTP or during routine cleaning and maintenance. It would be anticipated that the external dose from any discharge of radioisotopes to a worker who was simply present in the filtration room would be small due in part to spatial distances, attenuation effects of the metalwork of the filtration units and the temporal nature of any discharge pulse passing through the WTP. The apparent longer residence time of solid waste in the filtrate collector may provide exposures to workers at the Breivika WTP over longer time periods than any intermittent discharges of radioisotopes in the influent. Solid waste from the filtrate collector is further conveyed to a transport container in a separate room underneath the filtration room. Transport containers are exchanged when full (typically every 3 to 4 days), so further exposures from radioisotopes in the solid waste at this stage in the process are possible. Solid waste transported to the waste disposal site at Stormoen, Balsfjord is likely to contain residual activity from at least I-131, which may then result in exposures to workers at the waste disposal site.

Routine cleaning and maintenance of the filtration units may result in higher exposures to workers if they are carried out at the same time as when any pulse discharges of radioisotopes are within the WTP. Due to the apparent long residence time of solid waste in the filtrate collector, routine cleaning of the filtrate collector is more likely to lead to an increased chance of exposure from any discharges of radioisotopes. The type of exposure (external, skin contamination or inhalation) when carrying out routine cleaning or maintenance will depend on the procedure and the safety equipment employed by the worker. The degree of exposure will vary depending on the time exposed, proximity to the source and the activity concentration of radioisotopes in either the influent or the solid waste. From this study, it is not possible to predict maximum activity concentrations of I-131 (or any other radioisotope in use at UNN) in either the influent or the solid waste. However, external effective doses can be estimated using Microshield® radiation software for hypothetical exposures to a sphere (radius of 5.8 cm) of solid waste with a mass of 1 kg, a density of 1.2 g/cm<sup>3</sup> and the observed maximum activity concentrations of F-18, Tc-99m, I-123 and I-131 in solid waste. Estimated external effective dose rates at a distance of 10 cm and 1 m were low in each case and of the order of  $10^{-3} \,\mu \text{Sv/hr}$  or lower (Table 9). Summing the external effective dose rates for each individual radioisotope gives values of 0.013 μSv/hr at 10 cm and 0.00027 μSv/hr. The summed effective dose rate at 10 cm is similar to the average hourly dose rate reported for a worker at Breivika WTP in the assessment carried out by UNN, whereas the summed effective dose rate at 1 m is two orders of magnitude lower. Annual occupational doses (where a working year is equivalent to 2000 hours) based on

these summed dose rates would be at least 2 orders of magnitude lower than the individual dose limit for public exposure of  $1\,\mathrm{mSv/a}$ .

Table 9. Estimated effective external dose rates from a sphere<sup>1</sup> of solid waste with observed maximum activity concentrations of F-18, Tc-99m, I-123 and I-131.

	D // 6	μSν	ı/hr
	Bq/kg f.w	10 cm	1 m
F-18	772 ±23	4.43E-03	9.61E-05
Tc-99m	4507 ±315	4.90E-03	1.07E-04
I-123	5.9 ±1.0	8.48E-06	1.84E-07
I-131	1329 ±27	3.12E-03	6.77E-05

<sup>1 -</sup> mass of 1 kg, density 1.2 g/cm<sup>3</sup> and radius 5.8 cm.

#### 4 Conclusions

It is possible to establish a direct link between the use of I-131 at UNN and the observed activity concentrations of I-131 in sewage samples at Breivika WTP and in the marine environment around Tromsøya. Activity concentrations of I-131 can be detected in *F. vesiculosus* at distances of at least 10 km from the point of discharge from the Breivika WTP. For the treatment of a single patient at UNN with 6 GBq of I-131, activity concentrations of I-131 in *F. vesiculosus* can reach up to 400 Bq/kg f.w. at locations close to the point of discharge. However, activity concentrations of I-131 in *F. vesiculosus* at such locations can increase above 400 Bq/kg f.w. when the time between individual patients being treated with I-131 at UNN is less than one week. Beyond locations close to the point of discharge, all observed activity concentrations of I-131 in *F. vesiculosus* were low and below 10 Bq/kg f.w.. Mean effective and biological half lifes for I-131 in *F. vesiculosus* at locations close to the point of discharge were 3.6 days and 6.5 days, respectively. Committed effective doses to *F. vesiculosus* from I-131 were low and below expected noeffect levels. Recreational and ingestion exposure pathways to humans from I-131 in the marine environment are unlikely, with any potential dose far below international limits for the public. Estimated excretion rates for I-131 from patients at UNN based on composite sampling of influent and effluent at Breivika WTP were in reasonable agreement with previous in vivo studies.

Furthermore, it is possible to establish a direct link between the use of Tc-99m, I-123 and F-18 for diagnostic radiography purposes at UNN and the observation of activity concentrations of these same radioisotopes in sewage samples at Breivika WTP. Other than I-131, no other radioisotope in use by UNN was detected above detection limits in *F. vesiculosus*.

Hypothetical estimated effective dose rates at 10 cm and 1 m from solid waste with observed maximum activity concentrations of F-18, Tc-99m, I-123 and I-131 were comparable or lower than previously derived hourly dose rates for workers at Breivika WTP. It is likely that the main potential exposure pathways for workers at the Breivika WTP are when carrying out routine cleaning and maintenance of the filtration units and the filtrate collector, when such work practices coincide with the passage of radioisotope discharges.

The use of in situ detectors to directly monitor activity concentrations in the influent and higher frequency sampling of filtered solid waste would provide a more robust basis to understanding the magnitude of exposures to workers at the Breivika WTP from discharges of radioisotopes that are used at UNN.

## Acknowledgement

The authors would like to acknowledge the considerable help and assistance of Tord Bentzen, Water & Sewage Department, Tromsø Kommune and Arne Erikson and Anders Lund Eide, Nuclear Medicine Section, University Hospital of North Norway, Tromsø

#### References

- Audunson, T., Næser, H., 1975. Resipientundersøkelser ved Tromsø. Vann 4, 277-290.
- Brown, J.E., Kolstad, A.K., Brungot, A.L., Lind, B., Rudjord, A.L., Strand, P. and Føyn, L., 1999. Levels of <sup>99</sup>Tc in seawater and biota samples from Norwegian coastal waters and adjacent seas. Marine Pollution Bulletin 38(7), 560-571.
- Brown, J.E., Alfonso, B., Avila, R., Beresford, N.A., Copplestone, D.C., Prohl, G., Ulanovsky, A., 2008. The ERICA Tool. Journal of Environmental Radioactivity 99, 1371–1383.
- Carolan, J.V., Hughes, C.E., Hoffmann, E.L., 2011. Dose assessment for marine biota and humans from discharge of <sup>131</sup>I to the marine environment and uptake by algae in Sydney, Australia. Journal of Environmental Radioactivity 102(10), 953-963.
- Cooper, L.W., Beasley, T.M., Zhao, X.L., Soto, C., Vinogradova, K.L., Dunton, K.H., 1998. lodine-129 and plutonium isotopes in Arctic kelp as historical indicators of transport of nuclear fuel-reprocessing wastes from mid-to-high latitudes in the Atlantic Ocean. Marine Biology, 131(3), 391-399.
- Demir, M., Parlak, Y., Çavdar, I., Yeyin, N., Tanyildizi, H., Gümüşer, G., Sayit, E., Erees, S., Sayman, H., 2013. The evaluation of urine activity and external dose rate from patients receiving radioiodine therapy for thyroid cancer. Radiation Protection Dosimetry 156(1), 25-29.
- Druehl, L.D., Cackette, M., D'Auria, J.M., 1988. Geographical and temporal distribution of iodine-131 in the brown seaweed Fucus subsequent to the Chernobyl incident. Marine Biology 98(1), 125-129.
- FAO/WHO, 2016. Joint FAO/WHO Food Standards Programme, Codex Alimentarius Commission, Codex General Standard for Contaminants and Toxins in Food and Feed, Schedule 1 Radionuclides, Codex Standard 1931995, CAC, Rome.
- Garnier-Laplace, J., Copplestone, D., Gilbin, R., Alonzo, F., Ciffroy, P., Gilek, M., Agüero, A., Björk, M., Oughton, D.H., Jaworska, A., Larsson, C.M., Hingston, J., 2008. Issues and practices in the use of effects data from FREDERICA in the ERICA Integrated Approach. Journal of Environmental Radioactivity 99, 1474–1483.
- Guimaraes, J. R. D., Penna-Franca, E., 1985. <sup>137</sup>Cs, <sup>60</sup>Co and <sup>125</sup>I bioaccumulations by seaweeds from the Angra dos Reis nuclear plant region. Mar. Environ. Res. 16, 77–93
- Hevano, S., Islui, T., Nakamura, R., Matsuka, M., Koyanagi, T., 1983. Chemical forms of radioactive iodine in seawater and its effects upon marine organisms. Radioisotopes 33, 319-322.
- IAEA, 1996. International Atomic Energy Agency. International basic safety standards for protection against ionizing radiation and for the safety of radiation sources. Safety Series No. 115. IAEA, Vienna.
- IAEA, 2004. International Atomic Energy Agency. Sediment distribution coefficients and concentration factors for biota in the marine environment. Technical Reports Series No. 422. IAEA, Vienna.
- ICRP, 2004. International Commission on Radiological Protection. Release of Patients after Therapy with Unsealed Radionuclides. ICRP Publication 94. Annals of the ICRP 34(2).
- IPEM, 2018. Institute of Physics and Engineering in Medicine. Advice notice on excretion factors: the percentage of administered radioactivity released to sewer for routinely used radiopharmaceuticals.

# https://www.ipem.ac.uk/Portals/0/Excretion%20factors%20Sept%202018.pdf?ver=2018-10-03-150031-463

- Kelly, D.G., Mattson, K.M., McDonald, C., Nielsen, K.S., Weir, R.D. (2014). Environmental radionuclide monitoring of Canadian harbours: a decade of analyses in support of due diligence activities by the Royal Canadian Navy. Journal of Environmental Radioactivity, 138: 303-307.
- Klemperer, H. G., 1957. The accumulation of iodide by Fucus ceranoides. Biochemistry Journal 67, 381–390
- Mettler, F.A., Guiberteau, M.J. 2012. Radioactivity, Radionuclides, and Radiopharmaceuticals. In: Essentials of Nuclear Medicine Imaging (Sixth Edition), Elsevier pp.1-13.
- Pollard, D., Long, S., Hayden, E., Smith, V., Ryan, T.P., Dowdall, A., McGarry, A., Cunningham, J.D. (1996). Radioactivity monitoring of the Irish marine environment 1993 to 1995 (RPII--96/5). Ireland
- Statens Kartverk, 2008. Den Norske Los, Bind 6, Farvannsbekrivelse: Lødingen og Andenes Grense Jakobselv. Sjøkartverket, Stavanger.
- Sundell-Bergman, S., Avila, R., de la Cruz, I., Xu, S., Puhakainen, M., Heikkinene, T., Rahola, T., Hosseini, A., Nielsen, S.P., Sigurgeirsson, M., 2009. Assessing the impact of releases of radionuclides into sewage systems in urban environment-simulation, modelling and experimental studies LUCIA. Nordic Nuclear Safety Research, NKS-B report, No. 192, pp. 1-78.
- Vives i Batlle, J., Wilson, R.C., Watts, S.J., Jones, S.R., McDonald, P. and Vives-Lynch, S., 2008. Dynamic model for the assessment of radiological exposure to marine biota. Journal of Environmental Radioactivity 99(11), 1711-1730.

dsa@dsa.no +47 67 16 25 00 dsa.no

