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Geostatistical Methods Applied to Sampling Optimisation for the Temporal Monitoring of Technetium-99 in the Arctic Marine Environment

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Key words:
Arctic, marine, technetium-99, monitoring, optimisation.

Abstract:
This report details the demonstration of a spatial interpolation method towards the optimisation of temporal sampling for the determination of concentrations of technetium-99 in the Arctic marine environment. The impact of various sampling regimes on the quality of data produced using geostatistical interpolation is ascertained and the use of kriging error minimisation as a decision criterion is demonstrated. The use of an oceanographic model for the a priori determination of parameters relevant to the system used is also demonstrated.

Referanse:
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Emneord:
Arktis, marint, technetium-99, overvåking, optimisering

Resymé:
Denne rapport demonstrerer i detalj optimaliseringen av tidsmessig prøvetaking for å bestemme konsentrasjonen av Tc-99 i det arktiske miljøet. Innvirkningen av forskjellige prøvetakingssystemer hva angår kvalitet av data som blir brukt i geostatistisk interpolasjon er fastslått og bruken av matematisk minimering av feilkilder som et beslutningskriterium er demonstrert. Bruken av oceanografisk modell for bestemmelse av relevante parametre er også demonstrert.

Head of project: Tone Bergan.
Approved:

Per Strand, Director, Department for Emergency Preparedness and Environmental Radioactivity.

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Geostatistical Methods Applied to Sampling Optimisation for the Temporal Monitoring of Technetium-99 in the Arctic Marine Environment

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Content

1 Introduction

Monitoring of the marine environment for radioactivity remains one of the more expensive and labour intensive monitoring activities due to the large sample volumes required and the complex and lengthy analytical procedures required to measure current concentrations of radioactive contamination. Because of these factors some consideration must be given to the design of monitoring regimes to ensure effective and cost efficient sampling that can be defended on the basis of scientific rationale. Under the Norwegian Marine Monitoring Programme (RAME), the Norwegian Radiation Protection Authority (NRPA) and other institutions have for the past decade been conducting marine monitoring for a variety of radionuclides at a series of stations along the coast of Norway and at other relevant sites (Brown et al, 1999; 2002, Gerland et al, 2003). Such monitoring activities have resulted in the generation and maintenance of relatively long time-series relating to the levels of some radioactive contaminants in the marine environment. In recent years, the development of oceanographic models that describe the transport and occurrence of such contaminants in the marine environment have supplemented empirical data generated by monitoring programmes and provide an important tool for the optimisation and assessment of monitoring regimes.

This report details the results of an investigation of the applicability of advanced spatial analysis routines to the problem of optimising temporal sampling regimes for the monitoring of radioactive contaminants in the northern marine environment using ^{99}Tc as a case study. The results of such application are presented and discussed and the methods are appraised in relation to both empirically derived data and data provided by an oceanographic model. The results of the project are also used to analyse the efficacy of current monitoring programs for the monitoring of current and future of levels of this isotope in the Norwegian marine environment.

1.1 Technetium-99 (^{99}Tc)

^{99}Tc is a fission product of uranium and is formed during nuclear reactions by the beta decay of ^{99}Mo as presented in Figure 1.

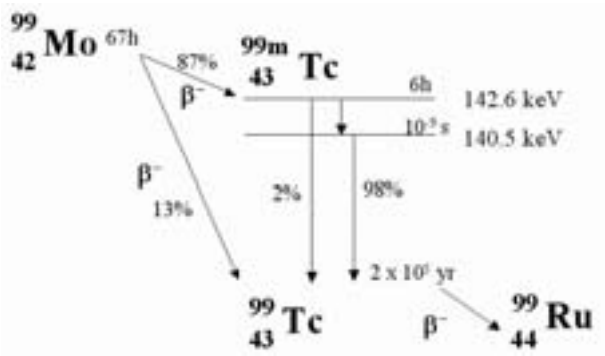

Figure 1. Decay scheme of ^{99}Mo

^{99}Tc, with a half-life of 213000 years and a specific activity of 630 kBq/mg, undergoes beta decay to form the stable isotope ^{99}Ru. Since its discovery, artificially created by the bombardment of molybdenum with deuterons (Perrier and Segre, 1947), 21 isotopes of the element have been prepared, with atomic mass numbers between 90 and 110 (Long and Sparkes, 1988). All known isotopes of technetium are radioactive and although miniscule amounts of ^{99}Tc have been found as a result of naturally occurring processes, namely the spontaneous fission of ^{238}U (Kenna and Kuroda, 1964), by far the greatest source of ^{99}Tc is the operation of nuclear reactors and associated facilities in the nuclear industry. An estimated 25-30 tonnes of the isotope have been produced worldwide with ca. 1% of that amount (150-200 TBq) having been released to the environment (Aarkrog et al., 1986, Beasley and Lorz, 1986) prior to the 1990's. Of the total amount released to the environment, ca.

85% has been released as a result of the activities of nuclear fuel reprocessing plants and the remaining ca. 15% has resulted from the testing of nuclear weapons (Dahlgaard, 1995). The long half-life of ^{99}Tc, its high environmental mobility as the poorly sorbed, chemically stable, anionic pertechnate species (TcO_4^-) and its infiltration of food chains as an analogue of sulphate (Cataldo et al., 1989, Bondietti and Francis, 1979) make the contamination of the environment with this isotope a matter of some concern and necessitates the establishment and maintenance of monitoring systems.

1.2 ^{99}Tc discharges from nuclear reprocessing facilities

Past and continued sources of ^{99}Tc to the Arctic marine environment are the nuclear fuel reprocessing facilities at Sellafield in the UK and Cap la Hague in France (Fig. 2). Both plants are engaged in the reprocessing of spent fuel from nuclear reactors.

Although some nations have chosen to store spent nuclear fuel from their reactors, the United Kingdom and France reprocess spent fuel produced by their own reactors and the reactors of some other countries. The reprocessing process involves dissolution of the nuclear fuel in acids with subsequent isolation of the reusable uranium and plutonium by a variety of chemical means. Other radioactive isotopes, formed during usage of the fuel, are liberated from the solid fuel by the dissolution process and are regarded in general as waste products. Such isotopes include ^{90}Sr, ^{137}Cs and ^{99}Tc. Much of this waste can be concentrated and stored on-shore for future disposal or continued storage. However a proportion of this waste is discharged to the oceans.

1.2.1 *Historical discharges of ^{99}Tc from the Sellafield and Cap la Hague facilities*

Discharges of ^{99}Tc from these facilities have varied in magnitude over the past decades (Figure 2). During the 1980's and into the early 1990's, the French facility at Cap la Hague dominated in terms of total ^{99}Tc discharged. In the period 1982-1991, Sellafield discharged a total of 42.4 TBq of ^{99}Tc (BNFL, 1982-1991) compared to a discharge of 102 TBq by the la Hague facility during the same period (Herrmann et al., 1995).

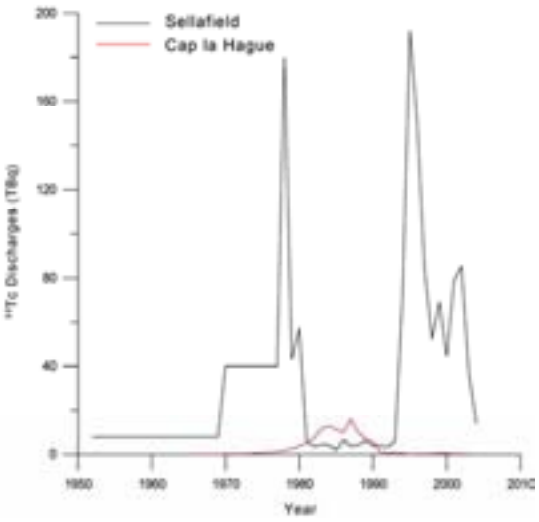

Figure 2. Historical discharges of ^{99}Tc from Sellafield and Cap la Hague.

In the period prior to the early 1980's, ^{99}Tc discharges were largely dominated by the Sellafield contribution although estimation of the magnitude of the discharges are complicated due to the fact that ^{99}Tc was not reported as an individual radionuclide prior to 1978. Leonard et al. (1997a) estimate discharges of ^{99}Tc between 1952 and 1968 to have been between 5 and 10 TBq per annum rising to 40 TBq per annum between 1970 and 1977. Between 1980 and 1994, ^{99}Tc was stored on-site at Sellafield whilst the Enhanced Actinide Removal Plant (EARP) was commissioned and discharges of ^{99}Tc were of the order of 4-6 TBq per annum. The EARP plant was designed to treat high level nuclear wastes arising from the operation of BNFL's Magnox reprocessing plant by

removing actinide and caesium isotopes from the waste. ^{99}Tc in the waste is present as the pertechnate ion and cannot easily be removed from the waste by the processes employed at the EARP plant (Busby et al., 1997). The net result of this fact has been that since 1994, ^{99}Tc discharges from the EARP plant at Sellafield have increased 50-fold from the discharge levels of the early 1990's, to a level of 150-190 TBq per annum in 1995 and 1996.

1.3 Transport of ^{99}Tc to the Norwegian Marine Environment

Leonard et al. (1997b) compared levels of ^{99}Tc in British coastal waters before and after the initiation of activities at the EARP plant and observed an increase in the levels of ^{99}Tc in the waters of the Irish Sea from 1-4 Bq/m^3 before operations at EARP commenced to in excess of 30 Bq/m^3 (northern Irish Sea) and 200 Bq/m^3 (off western Scotland) after EARP began processing nuclear waste at the Sellafield site. Such observations indicated that the ^{99}Tc had migrated to the northern reaches of the Irish Sea in a relatively short period of time (approx. 3 months). The ^{99}Tc signal was then lost due to inflows of water from the Atlantic Ocean but reappeared in the North Sea having taken ca. 9 months to traverse the distance between the point of discharge at Cumbria in the UK and the waters off the northern coast of Scotland.

The findings of Brown et al. (1998) indicated that the ^{99}Tc from the increased Sellafield discharges beginning in 1994 had reached Norwegian coastal waters some time before November 1996 and could be detected in waters off Northern Norway by December 1997. Tc-99 is transported to Norwegian waters by the Norwegian Coastal Current via the North Sea. This current continues along the Norwegian coast before flowing further north as the West Spitsbergen Current, transporting any contaminant load into the Norwegian Arctic marine environment (Figure 3).

The most recent data regarding levels of ^{99}Tc in the waters and biota off mainland Northern Norway indicate an average level of 1.25 $\pm$ 0.33 Bq/m^3 in seawater and 321 $\pm$ 68 Bq/m^3 in seaweed (d.w.). These values continue to demonstrate the contemporary elevated levels of this isotope in the Norwegian marine environment and provide the impetus for the maintenance of current monitoring activities pertinent to the occurrence of this isotope in this region.

Figure 3. Oceanic currents involved in the transport of ^{99}Tc to the northern marine environment.

1.4 Monitoring of ^{99}Tc in the Norwegian Marine Environment

In 1999, sampling and analysis of waters and biota along the Norwegian coast by the Norwegian Radiation Protection Authority increased under the new marine monitoring programme (RAME) funded by the Norwegian Ministry of the Environment, the primary objectives of which are to monitor and document concentrations of and trends in radionuclide contamination in Norwegian waters. Previously, data have also been generated regarding levels of radioactivity within Norwegian waters by various institutes and organisations as part of such institutes monitoring/research activities. Such data used

in this report were generated by the Institute for Energy Technology (IFE), Norway as a result of monitoring activities conducted at Utsira in Southern Norway.

Monitoring within the RAME programme is conducted via regular sampling at a number of sites within the Norwegian Arctic waters, participation in cruises and sampling during field expeditions on Svalbard. The aim of these activities is to obtain information regarding the occurrence and behaviour of ^{99}Tc and other radionuclides in the Norwegian Arctic marine environment.

Although recent years have witnessed reductions in the amounts of ^{99}Tc discharged from European reprocessing installations, the long half-life of this isotope and its occurrence in food chains necessitates monitoring of levels of the isotope for some time to come.

2 Geostatistical Methods

The method selected in this study is a suite of procedures commonly termed "geostatistics" in deference to their original development within the field of geological sciences. Empirically described by Krige (1951) and theoretically grounded by Matheron (1963) some years later, geostatistical methods were initially exclusively employed in the field for which they were developed which was ore reserve estimation. In the past two decades however the methods have been increasingly applied to the problem of pollution assessment and the mapping of environmental variables, the surge in interest and their growing acceptance being reflected in the increase in the amount of literature on the application of geostatistics in the field of environmental monitoring. Geostatistical methods are primarily concerned with the estimation of the value of a variable at a particular point (or within a certain volume) in space or time based on information provided by actual data for sampled points and certain characteristics of the relationship exhibited by different points for values of the variable. The outcome of the process is typically a set of estimates for unsampled points and an associated uncertainty with those estimates.

Traditional methods of estimation and interpolation have relied on two distinct approaches: the "area of influence" method and classical statistical treatments. The former utilises both the spatial position of the known samples and the value of the variable at those points, the latter adheres to classical statistical theory by assuming the known sample values to be random observations drawn from a given population, no regard being given to the spatial location of the samples. Both methods have proven to be far from ideal for a number of reasons. The "area of influence" method, typically demonstrated in polygonal estimation, is unable to produce confidence limits or a measure of the reliability of the produced estimates. Although classical statistics are in a position to produce such uncertainty values, the spatial or temporal location of the known samples is ignored in the estimation procedure, an omission which was deemed undesirable by the geologists using the techniques for the purpose of reserve valuation, ultimately leading to the development of geostatistical methods and the underlying theoretical principles – the Theory of Regionalised Variables.

2.1 Theory of Regionalised Variables

A fundamental concept underling geostatistical theory and practice is the concept of the "Regionalised Variable". A random variable (RV), being a variable that varies in a probabilistic manner between individual samples, differs from a regionalised variable (ReV) in that the latter is a random variable that varies in value according to its location within

some predetermined area or time, i.e. an ore body, or survey region. Journel and Huybregts (1978) define the two components of a regionalised variable as:

I. an observation of the variable at point or time w_i within the greater area or time w, is a realisation of a random variable $Z(w_i)$ for the point w_i. This component is random in that the set of random variables for every point confined by w is a random function,

II. the random variables for two locations, w_i and w_{i+h} (separated by a vector or time h) are not considered independent. i.e. $Z(w_i)$ and $Z(w_{i+h})$ are spatially or temporally correlated.

The description of regionalized variables provided above is a mathematical expression of the phenomena intuitively recognised by many practitioners of environmental monitoring, that samples separated by great distances or time spans are usually more dissimilar than samples separated by smaller distances or time spans. Such a phenomenon is not recognised within classical statistical approaches but is inherently incorporated within the geostatistical methods and is one that has led to its increasing implementation in the field of environmental monitoring. The characteristics of regionalised variables as described above can be incorporated into the concept of a "Random Function" (RF). At each point w_i in two dimensional space, or time t_i in the time dimension, the observed data value is considered to be a realisation of a certain random variable $Z(t_i)$ (or $Z(t_i)$) defined for point w_i or time t_i. The set of all random variables $\{Z(x \text{ or } t_i)\}$; x or t_i in the space or time domain is a random function or RF. Given that this report concerns itself with the temporal dimension, the discussion following will only concern itself with t_i. For a given pair of points t_i and t_{i+h} separated by time h, the corresponding ReV's $Z(t_i)$ and $Z(t_{i+h})$ are not generally independent. This non-independence is the structured component of the ReV. The total set of observations $z(t_i)$ at times t_i over the whole period is considered to be the realisation of the RF. As only a single realisation is usually available some assumptions must be made as to the 2nd order stationarity (the time-invariance of the data's' probability distribution) of the data so that data for different times can be viewed as multiple realisations of the same ReV. The assumptions as they relate to geostatistical methods and practice are known as the Intrinsic Hypothesis.

2.2 The Intrinsic Hypothesis

Geostatistical methods involve the analysis of the spatial or temporal correlation of the variable in question and the production of estimates for unsampled locations or times based on the extent and nature of the spatial or temporal correlation. The former is achieved using a technique known as semi-variogram analysis, the latter employing an estimation procedure commonly known as kriging. Both these methods rely on the data set being used fulfilling the underlying intrinsic hypothesis. This hypothesis is generally satisfied if:

I. The expectation value of the difference between two points, t_i and t_{i+h}, separated by time h, should be zero,

II. The variance of the differences between points separated by time h is only a function of the magnitude of h or alternatively;

$$Var[Z(t_{i+h}) - Z(t_i)] = 2\gamma h \qquad [1]$$

where γ is the semi-variogram value which is introduced later.

The initial assumption of the intrinsic hypothesis is best explained with the use of an example. A number of samples, all separated by a time h, are analysed for a variable denoted q. If the difference in the value of q exhibited by samples separated by h is only a function of h, then the distribution of the differences depends on h and hence the mean and variance of the distribution

also depend on *h*. The mean difference, *a*(t)*, between points (denoted *w*) can be established by:

$$a^*(h) = \sum [q(w) - (q(w+h)] \; 1/n \qquad [2]$$

where *n* is the number of points.

The term *a*(h)* can also be deemed the 'expected' difference between samples separated by the time, *h*. If this value is zero then no difference is expected between samples separated by *h*. In practical terms this means that for a period determined by the magnitude of *h*, all samples should exhibit similar values for the variable *q*. If this is not true then a trend is said to exist within the area, or the expected difference between two points does not equal zero.

The second part of the intrinsic hypothesis concerns itself with the concept of "stationarity of difference". If the above example is implemented again, the variance of the distribution of the differences between samples separated by a time h may be given by $2\gamma(h)$, also called the variogram value. This value should only be a function of the magnitude of *h* and not the point within the study period at which *h* occurs. If the variogram value for points separated by a time *h* relies on the location within the time series for which that value is calculated, the second requirement of the intrinsic hypothesis is not fulfilled.

The extent to which the fulfilment of the conditions of the intrinsic hypothesis affects the implementation of geostatistical procedures remains unclear from the examples provided in the literature. Although many researchers include a statement of the intrinsic hypothesis in their articles (McBratney and Laslett, 1993, Einax and Soldt, 1995), none give any indication of their having tested their data for fulfilment of the conditions. The presence of a trend in a data set is often given as a reason for the implementation of advanced data modifications to remove or model the trend (Flatman and Yfantis, 1984). Clarke (1979), in one of the standard texts on the subject, states that trends in a data set are not a problem insofar as they may manifest themselves at distances which are great enough that they do not pose a problem for local estimation (ie. at distances greater than the range of correlation).

Clarke also states that there is no statistical test for stationarity, and that stationarity within a data set may only be assumed and not proven. Stationarity is a function of the random component of the spatial structure, not the data itself, and a test would therefore require multiple realisations of the random function, a feat not possible in practice. Typically, a state of "quasi-stationarity" is deemed acceptable where stationarity is present locally or over the area or time for which estimation will be made.

2.3 The Semi-variogram

The extent and form of the spatial, or in this case temporal, correlation of the data set is investigated using semi-variogram analysis. The semi-variogram is a graphical representation of how the similarity between variable values varies as a function of the distance (and direction) or time separating them. The theoretical semi-variogram is a plot of one half of the variance of the differences in variable values (y axis) as a function of the distance, or "lag", separating pairs of points (x axis), the general equation for the semi-variance being:

$$\gamma(h) = \frac{1}{2n(h)} \sum_{i=1}^{n-h} \left[z(x_{i+h}) - z(x_i) \right]^2 \qquad [3]$$

where $\gamma(h)$ is the semi-variance, $n(h)$ is the number of points separated by the time or distance *h* (the "lag") and $[z(x_{i+h}) - z(x_i)]$ is the difference between the values of variables separated by the lag *h*.

As only a limited number of pairs are available in a practical study, an *experimental* semi-

variogram is plotted using the available data and a theoretical model is fitted to the resulting plot. The curve fitted to the data can be seen to usually consist of two distinct parts, an initial rising segment and a second, level region (Fig.4). The point at which the curve levels off is used to calculate both the *sill* (y axis) and the *range of correlation* (x axis).

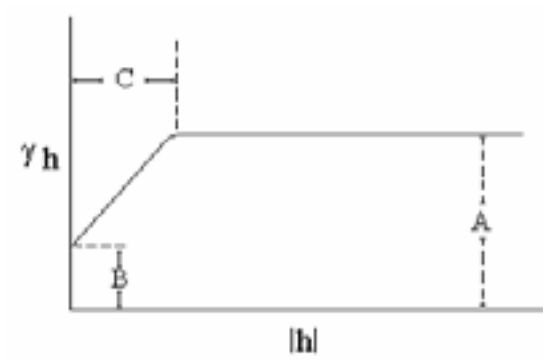

Figure 4. Schematic representation of a typical semi-variogram structure.

The sill is the maximum semi-variance exhibited by the data set and the range of correlation is the lag (or separation distance or time) at which the sill value is reached. Pairs of points separated by a distance or time greater than the range of correlation are considered to be spatially or temporally uncorrelated. A sample can be taken as representative of an area or time defined by the range of correlation. The range of correlation provides a mathematical means of measuring the "area of influence" described in other estimation methods. The semi-variance value indicated by the sill can be divided into two components, B and A-B in Fig. The "nugget effect", often-denoted Co, (B in Fig.4) represents the random variance in the data. The semi-variance, represented by A - B in Figure 4 is the structured component of the data set's variance.

The experimental semi-variogram only provides information on the data set used to construct the plot. In practice it is often not possible to compute semi-variances for the exact lags plotted in the semi-variogram, especially if the data has not been taken from a regular grid. The specification of a lag "tolerance" is therefore common practice. As an example, for a defined lag of "x" with a lag tolerance of "+\- 2" then pairs of points of "x+1" and "x-1" would both be included in the set used for calculation of the lag.

As it is not possible to sample every possible point and in order to describe the entire period (and lag distances for which a semi-variogram value has not been computed), it is necessary to fit a mathematical model to the data to produce the theoretical semi-variogram. A number of models are frequently used to describe the theoretical semi-variogram and some of these are briefly described below.

The most commonly used model in geostatistics, the spherical model, is indicative of a high degree of spatial or temporal continuity. It is linear near the origin before flattening out as it approaches the sill at the range of correlation. It is described by the equation:

$$\gamma(h) = 1.5h/a - 0.5(h/a)^3 \qquad [4]$$

where a is the range of correlation and h is the lag distance or time.

The exponential model reaches the sill asymptotically, being linear near the origin, and may be described by the equation:

$$\gamma(h) = 1 - exp(-3h/a) \qquad [5]$$

Gaussian models are used to model extremely continuous variables. Exhibiting parabolic behaviour near the origin, the Gaussian model reaches the sill asymptotically, the range being accepted as the point at which the curve reaches 95% of the sill value:

$$\gamma(h) = 1 - exp(-3h^2/a^2) \qquad [6]$$

Power models are a collection of models described by a general equation:

$$\gamma(h) = h^n + C_o \qquad [7]$$

where $0 < n < 2$, h is the lag distance and C_o is the nugget of the semi-variogram.

A significant feature of many data sets is a state of anisotropy. A data set is said to be isotropic if the properties of the semi-variogram remain constant for all the directional variograms. A directional semi-variogram is a semi-variogram plotted for one direction only, i.e. N-S or E-W. If the range of correlation or sill of a semi-variogram model differs between directional components, then it is said to be anisotropic. If the only difference between directional semi-variograms is the length of a, the range, then the anisotropy is geometric. Zonal anisotropy occurs when the directional semi-variograms differ in relation to the value of the sill. Correction for geometric anisotropy involves incorporating the different range lengths into an ellipsoid describing the zone of influence around any sample point, as opposed to the circle of influence exhibited by points in an isotropic data set. Zonal anisotropies cannot be removed in this way and the different semi-variogram structures for the various directions must be accounted for in any subsequent estimation procedures. Given that a temporal series only extends in one direction, the concept of anisotropy in the case presented in this report is therefore redundant.

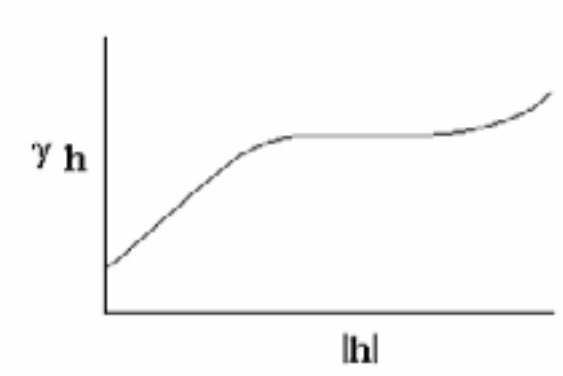

Figure 5. Semi-variogram fitted with Gaussian model displaying evidence of trend at longer lag periods.

A second significant feature of many semi-variograms is a parabolic ascension of the semi-variogram at some point an example of this being shown in Figure 5. Clarke (1979) demonstrates the use of this feature in the identification of trends within a data set and concludes that as long as the trend does not occur near the range of correlation then it may safely be ignored. This view is supported by Royle and Hosgit (1974), who cite an example of a sand and gravel deposit which displayed a parabolic upward curve, this feature being used as proof of the existence of a trend within the data. These workers also conclude that unless the parabolic nature of the semi-variogram occurs before or near the range of correlation it poses no difficulty to further stages in the geostatistical process if estimation is restricted to distances or times which are not affected by the trend. In some cases, insufficient point-pairs at greater lag distances result in unreliable estimates of the semi-variogram value which may produce a trend like effect for large lags. It is therefore common practice to limit the semi-variogram to lag distances that are only half the maximum lag exhibited by the data set in question.

2.4 Semi-variogram Analysis: Lognormal Data Sets

The treatment of data sets with log-normal distributions in the construction and modelling of semi-variograms has been the subject of much discussion. Gilbert and Simpson (1983) and Litaor (1995) both recommend the logarithmic transformation of lognormal distributions in order to better approximate a normal distribution, both presenting un-substantiated evidence that geostatistical procedures perform better on normally distributed data. McBratney and Laslett (1993) hypothesise that the nugget (the random component of the spatial correlation) may be over emphasised in semi-variograms of lognormal data due to the high values at the upper end of the distribution being confused with 'random noise' in the semi-variogram structure.

2.5 Geostatistical Estimation: Kriging

The estimation method known as kriging employs a weighted moving average interpolation procedure. Weights are assigned to known samples, the assignation of weights being governed by the information provided in the semi-variogram. Generally the closest neighbouring samples are attributed the greatest weights although in a data set exhibiting geometric anisotropy, a sample a distance away from an unknown point may be attributed a heavier weighting than a nearer sample depending on the ranges of influence in the two directions of the anisotropy. Kriging may either produce estimates for points in space or time (punctual) or for three dimensional volumes (block kriging). Block kriging is usually implemented in the estimation of ore bodies or similar phenomena, punctual kriging being used in environmental studies. The function for point kriging is:

$$z^*(t_0) = \sum_{i=1}^{n} \lambda_j z(t_j) \qquad [8]$$

where $z^*(t_0)$ is the estimate value at the unknown location or time t_0, λ_j being the weight assigned to the known sample z at (t_j). The weights are assigned according to the following equation,

$$\sum_{j=1}^{n} \lambda_i \gamma(t_i, t_j) + \mu = \gamma(t_j, t_0) \qquad [9]$$

$$i = 1, 2, ..., n.$$

subject to a number of constraints:

(t_i, t_j) being the time separating points t_i and t_j, (t_j, t_o) being the time between the unknown point t_o and the known point t_i, μ being a Lagrange multiplier. A distortion free distribution of weights is forced by using the following constraint:

$$\sum_{i=1}^{n} \lambda_i = 1 \qquad [10]$$

One of the most often cited advantages of kriging as an estimation method is the production of a measure of the error or uncertainty associated with the estimates produced by the kriging process and it is this parameter that shall be utilised in this study. As such it is worth taking a closer look at the parameter and the factors that contribute to it.

The production of this indicator of the estimate reliability (known as the "kriging standard deviation") is achieved as demonstrated by the following example. A time t, for which we wish to estimate the value of the variable q, is surrounded by many other points for which the value of q is known. A semi-variogram for the data set which contains these points has been calculated and a theoretical model has been fitted. When the value of q at t is estimated an error is incurred:

$$error = q_t - q_t^*$$

where q_t is the actual value of the variable q at t and q_t^* is the estimate of the value of the variable q at t. If the estimating procedure is unbiased (i.e. no trend present and a normal distribution of errors) then repeated estimations of q_t^* will have an average error of 0. The spread, or standard deviation, of these errors gives an indication of the reliability of the estimating procedure for the time t. For the purpose of this example, the estimation error variance will be denoted *'e*var'*. The variance of the errors can be calculated (theoretically) as the mean squared deviation from the mean error. This is equal to:

$$variance\ of\ errors = \frac{(error - mean\ error)^2}{n}$$

n being the number of estimations performed. However, as the estimating procedure is unbiased, the mean error will be equal to 0, the variance of the errors reducing to:

$$\text{variance of errors} = \frac{(q_t - q_t^*)^2}{n}$$

Practically, the kriging variance is given by:

$$\sigma_k^2 = \sum_{k=1}^{n} \lambda_i \, \gamma(|t_i - t_0|) + \mu \qquad [11]$$

As the value n represents the number of samples (whose values of q are known) used to estimate q_t^*, the semi-variogram value will be known for each of the samples that constitute the set n, as the time that separates t from each of the samples is already known (the lag value h). As the semi-variogram value is half the variance of the differences in the variable q exhibited by pairs of samples separated by a time h, multiplication of the semi-variogram value by 2 will give a measure of the reliability associated with an estimate made based on a sample a time h away from the unknown point. Although the above example presents the situation pertaining to kriging standard deviations a little simplistically, a consideration of the issue will indicate that the standard deviations depend on two factors only: the spacing of the known data points and the semi-variogram value. It is this feature of the process that makes it amenable to the design of sampling schemes.

The discussion of geostatistical theory presented in the previous section has, by necessity, maintained a relatively low level of mathematics. More detailed presentation of the mathematical foundation of geostatistics and the associated procedures may be found in Journel and Huybregts (1978), David (1977) and Krige (1951).

2.6 Sampling Design for Geostatistical Surveys

The design of sampling schemes for geostatistical surveys has received some attention in the literature, the vast majority of this literature pertaining to two-dimensional (time or location vs. a measured parameter) data sets although some of the findings do provide insight into how the techniques may be applied to temporal environmental series. Wang and Qi (1998), using artificially generated sample sets, ascertained that a regular grid system exhibited the best estimation performance. McBratney and Laslett (1993) recommend a random approach to sampling when the data displays a short range of correlation, nearest neighbour sites close to the unknown having strong spatial correlation. Geostatistical procedures appear well suited to the spatial analysis of soil properties and constituents for a number of reasons. Many soil properties, including radionuclide levels, are controlled by the underlying geology. Such parameters, in contrast with those such as ^{137}Cs soil levels (governed primarily by meteorological variables), are more likely to be spatially correlated, nearby samples being more similar than samples separated by large distances. This finding is of some consequence in the appraisal of the methods for temporal analysis of ^{99}Tc or other marine radiological contaminants as the occurrence of the nuclides is governed by underlying processes, most importantly, oceanographic processes. The application of geostatistics to radiological surveys is mostly confined to the terrestrial environment and for analysing distributions in space rather than time. Gilbert and Simpson (1983) investigated the application of geostatistical procedures for the spatial analysis of fallout patterns around nuclear test sites in the U.S., Badr *et al* (1996) utilising semi-variogram analysis to confirm geological control over soil gas radon concentrations. Geostatistical procedures have also been applied to the analysis of a number of other environmental parameters including soil pH (Yost et al, 1982), conductivity (Russo and Bressler, 1981), sodium levels (Burgess and Webster, 1980) and sand content (Vauclin et al, 1982).

Srivastava and Isaaks (1989) established that geostatistical methods performed better than other methods of estimation for a number of parameters and hypothesised that this was due

to the implementation of a customised statistical measure of distance rather than a geometrical description. Litaor (1995) observed, in comparing isopleth maps produced by geostatistics with those produced by a least squares method, that the kriging method eliminated some features visible in the other maps. He ascribed the differences to the smoothing effect of the process, kriging typically overestimating low values and underestimating high ones. The assessment of the performance of kriging in relation to other methods has been routinely performed in the mining industry. Barnes (1977) compared geostatistics to a polygonal and inverse distance procedures for ore evaluation, the geostatistical method performing favourably. Knudsen *et al* (1978) contrasted geostatistics with more conventional estimation methods, kriging outperforming the other methods over the chosen parameters. McBratney and Laslett (1993) point out that the implementation of geostatistics is often justified on the basis that the values in a data set are more similar the closer they are in space. Their argument is that other estimation procedures can highlight and use this feature and that an empirical comparison of geostatistics in relation to other methods has not been carried out in an environmental setting. Dowdall and O'Dea (1999) analysed the performance of geostatistical methods over conventional interpolators for natural radionuclides in a terrestrial environment and confirmed that the former outperformed the latter for a number of parameters.

Although geostatistical procedures appear to be gaining acceptance in the field of environmental monitoring, its only application to radionuclides appears to have been within the area of spatial analysis of natural and anthropogenic radionuclides in the terrestrial environment. There is little or no indication in the available literature of its having being applied to the monitoring of radionuclides in the marine environment or the temporal dimension. While geostatistical methods appear to offer a number of significant advantages over more conventional estimation techniques and seem to offer potential regarding the analysis of the temporal distribution of radionuclides in the marine environment, a number of points are worth considering. The application of geostatistics remains relatively new in the field of environmental monitoring and has not been implemented long enough to gain wide acceptance.

Geostatistical methods are quite general in relation to scale, examples in the literature have ranged from centimetres to kilometres but it should be realised that as the spacing between samples increases, the advantages of treating a variable as regionalised become less and geostatistical results begin to approach those of classical statistics. A large number of unresolved issues appear to exist in the field. There remains no true measure of the goodness of fit of any theoretical model applied to the raw semi-variogram, the most appropriate model often being chosen on the basis of trial and error. The assumption that the applied model describes spatial variability over the full region or period must always be made. Disagreement appears to exist on the correct treatment of log normally distributed variables and data sets exhibiting trends nor can the assumptions of the intrinsic hypothesis be verified in any meaningful way for any data set.

Despite these caveats, the implementation of a geostatistical procedure, after a meaningful and thorough examination of a data set, can offer advantages over other estimation procedures. At the very least, the use of such methods allows for a more in-depth analysis of a surveys' results than would be provided through the blind use of some of the more conventional methods. The computational requirements of geostatistics and the lengthy analysis involved, in combination with the problems listed above may serve to deter researchers in the environmental field from the adoption of such methods. Possible evidence for this is the large amount of published material in the statistical and mathematical journals and the relative lack of material in the earth science and environmental area. When geostatistics are

used, the implementation often appears to suffer due to either a lack of exploratory analysis of the data or via the use of commercial software with little regard for the quality of the final outputs. The use of geostatistical methods in the design of sampling schemes has again been limited to the spatial terrestrial environment although there is no evidence to suggest that it cannot be applied to temporal problems.

2.7 Optimisation of Sampling Regimes

The monitoring of marine radioactivity is both labour intensive and relatively expensive due mainly to the levels currently being observed in the marine environment, the large samples that must be taken and the lengthy analytical procedures required to conduct the analysis. This is particularly true for ^{99}Tc analysis with respect to both sampling and analysis. Because of these constraints, it is worth devoting some time to the design of a sampling plan and while obvious considerations include available resources and the purpose of the monitoring activity, some consideration must be given to the statistical or mathematical reasoning behind the chosen plan.

2.7.1 Power Analysis

The approach most often adopted in the design of temporal monitoring programs, in combination with practical considerations of local conditions, limitations and possibilities, is the Power Analysis. The concepts of statistical power are covered in detail in a number of texts (Kraemer and Thiemann 1987, Cohen 1988, Lipsey 1990; Muller and Benignus 1992). Briefly, the power of a test is the probability of rejecting the null hypothesis given that the alternative hypothesis is true. Power depends on the type of test, increases with increasing sample size or frequency, effect size, and higher a -level, and declines with increasing sampling variance. Effect size is the difference between the null and alternative hypotheses, and can be measured either using raw or standardized values. Raw measures, such as the difference between means or slope in a regression analysis, are closer to the measurements that researchers take and so are easier to visualize and interpret. Standardized measures, such as d-values or correlation coefficients, are dimensionless and incorporate the sampling variance implicitly, removing the need to specify variance when calculating power.

Although a full discourse on the theory and use of Power Analysis is beyond the scope of this report, the basic concepts are introduced below. A statistical hypothesis test provides a measure of the probability of a result occurring if the null hypothesis is false. If the probability is lower than a pre-specified value (alpha, usually 0.05), it is rejected. The ability to reject this null hypothesis depends upon:

1. Alpha (α): Usually set to be 0.05. This being the probability of a type I error, that is the probability of rejecting the null hypothesis given that that the null hypothesis is true. As an example, it is the probability of deciding that an increase of ^{99}Tc in the marine environment has been found when it has not actually occurred.
2. Sample frequency: A higher sampling frequency leads to more accurate parameter estimates, which leads to a greater ability to determine changes in the parameter of interest. The more often the observation is made, the greater the chance of observing the change.
3. Effect Size: The size of the change in the population. The larger the change, the easier it is to find.

However, the argument that the above statements may not be strictly correct is propounded by Cohen (1988) who points out that "*all null hypotheses, at least in their 2-tailed forms, are false*." Essentially this means that the change that is being sought or checked for (eg.

increase in ^{99}Tc concentrations) will always be present – it might just be present in such small quantities that it is below the level of change deemed worthy of determining by the designers of the monitoring program.

Power analysis permits the designer of the program to make sure that the sampling frequency is enough to observe the change with a certain probability and if that change is of a level of significance to be interesting. The size of the change being sought is known as the "effect size" (a doubling of ^{99}Tc concentrations for example). Several methods exist for deciding what effect size we would be interested in. Different statistical tests have different effect sizes developed for them, however the general principle is the same.

Ideally, power analysis is carried out *a priori*, which is during the design stage of the monitoring program. Given the three factors α, sample size and effect size, a fourth variable can be calculated, denoted β. Where α is the probability of a type I error (i.e. rejection of a correct null hypothesis) β is the probability of a type II error (acceptance of a false null hypothesis). The probability of correctly accepting the null hypothesis is equal to $1-\alpha$, which is fixed, the probability of incorrectly rejecting the null hypothesis is β. The probability of correctly rejecting the null hypothesis is equal to $1 - \beta$, which is called "power". The power of a test refers to its ability to detect what it is being looked for. A power analysis program can be used to determine power given the values of α, sample size or frequency and effect size. If the power is deemed to be insufficient, steps can be taken to increase the power, in this case via an adjustment of the sampling frequency.

Power calculations can be done using the tables or charts provided in many articles and texts (e.g., Kraemer and Thiemann 1987, Cohen 1988, Lipsey 1990). However these often require some hand calculations before they can be used, including interpolation between the tabled values, and can give inaccurate results in some situations. Computer software has the potential to make power analysis more accurate, interactive and easy to perform.

2.7.2 Kriging Error Minimisation

As introduced earlier, the parameter known as the kriging standard deviation has the potential to offer a means of optimising a sampling scheme assuming that a sampling scheme is required to provide estimates of levels of a contaminant at specific intervals over a certain time period. It is intended to take a certain number of samples and estimate the values for the desired time points using kriging. The desired uncertainty in the estimates is known and a semi-variogram has been determined. As the uncertainty in the kriged estimate is only a function of the variograms parameters (known already) and the sampling spacing, various sampling schemes can be tested to observe their effect on the kriging uncertainty. As a number of points will be estimated, there will be a range of uncertainties and some statistic relating to the distribution of the uncertainties must be selected as the governing criterion. The effect of sampling frequency on the magnitude of the chosen criterion can then be tested *a priori* and a sampling scheme selected. Changes in circumstance regarding the monitoring of the contaminant can then be easily accommodated and the design modified as required. It is this process that has been investigated in this study and for which results are reported.

3 The Data Sets

The data sets used in this study consisted of three radiometric environmental time series. Two of these consisted of ^{99}Tc values for seawater samples and the third consisted of ^{99}Tc values for seaweed samples. The time series were drawn from two locations along the Norwegian Coast. The first of these, Hillesøy (Fig. 6), is located in northern Norway (69.63N 17.95E). Seawater samples from this location have been taken by the Norwegian Radiation Protection Authority as part of its marine monitoring program since July 1997 and the time series used in this study begins on the 23rd of July 1997 and ends on the 30th of May 2003. As part of that program seaweed samples are also drawn from the same location and the time series utilised stretches from the 23rd of July 1997 to the 27th of February 2003. In both cases the sampling interval is approximately 30 days. The second seawater time series was drawn from Utsira (Fig. 6) in southern Norway (59.32N 4.90E) as part of monitoring activities conducted by IFE.

Figure 6. Locations of Hillesoy and Utsira

time series is also approximately 30 days. Summary statistics for all series are provided in Table 1 and representations of all the series are provided in Fig.'s 7 - 9. ^{99}Tc was measured in 50 l water samples and 10 g seaweed samples using a radiochemical procedure described by Chen et al (2001). Uncertainties for seawater measurements are of the order of 15% or greater, the uncertainty for seaweed being lower due to the greater analytical signal presented to the detector.

Both of the Hillesøy data sets pass the Kolmogorov-Smirnov (K-S) test for normality (Hillesøy water: $p = 0.8149$, Hillesøy seaweed: $p = 0.4577$), the Utsira data being best described by a lognormal distribution as reflected in its skewness and kurtosis coefficients. The Utsira set is strongly affected by a number of high data points at the beginning of the series. Evidence of trend is visible in the Hillesøy and Utsira seawater series, the evidence being less apparent for the seaweed series.

	Utsira	Hillesoy	
	^{99}Tc in seawater (Bq/m^3)	^{99}Tc in seawater (Bq/m^3)	^{99}Tc in seaweed (Bq/kg)
n	71	65	57
Mean	0.81	1.09	245.9
Std. Deviation	0.54	0.37	92.1
Minimum	0.2686	0.37	0.37
25th percentile	0.415225	0.85	197
Median	0.6005	1.05	257.5
75th percentile	1.136	1.285	309.25
Maximum	3.081	2	423
Skew	1.64	0.412	-0.156

Table 1. Summary statistics for both the Utsira and Hillesoy data sets.

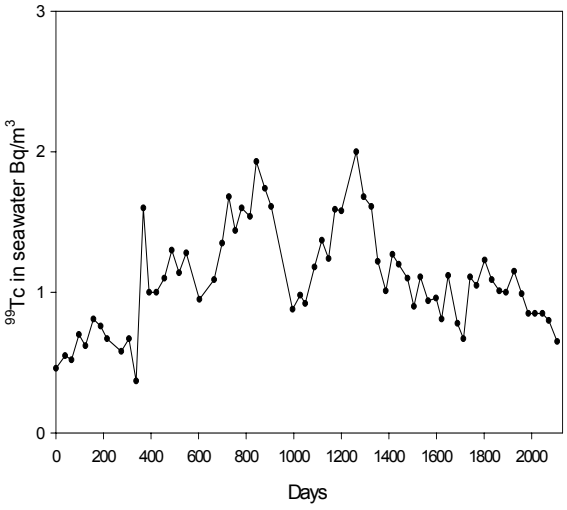

Figure 7 .Time series of ^{99}Tc in seawater from Hillesøy (start date: 23rd of July 1997).

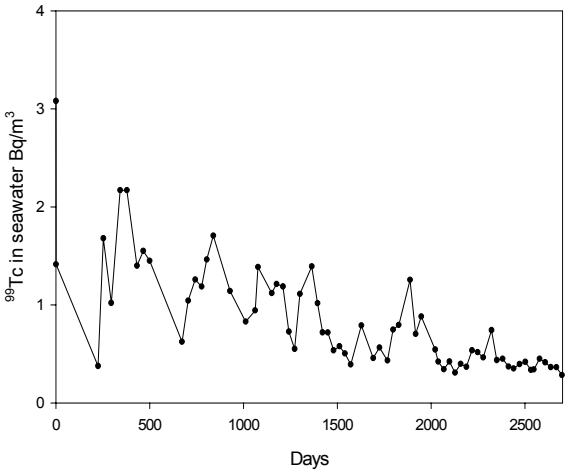

Figure 8. Time series for ^{99}Tc in seawater from Utsira (start date: 4th of July 1985).

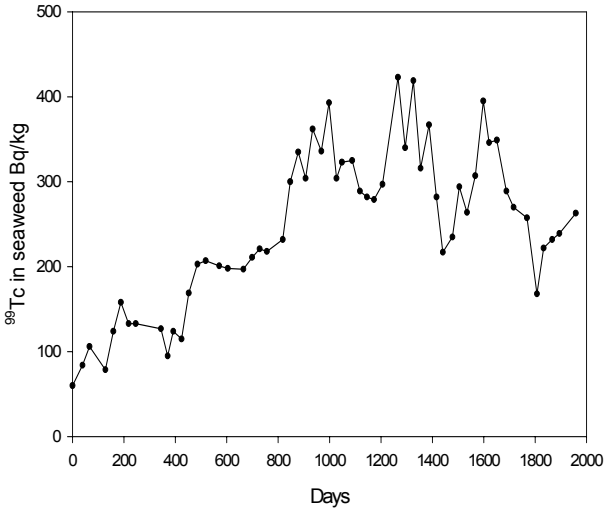

Figure 9. Time series for ^{99}Tc in seaweed from Hillesøy (start date: 13th of July 1997).

3.1 Semi-variography

Semi-variograms for the three data series being studied were constructed using Variowin 2.2 (Pannatier, 1996). Maximum lag periods in all cases were limited to half the maximum temporal separation of the data points to ensure enough points for calculation at the larger lag distances. The experimental semi-variograms for all three time series display strong temporal correlation (Fig.'s 10 – 12), the semi-variance for groups of samples separated by the shorter distances being less than that exhibited for groups separated by longer periods. For the Utsira time series the data was not transformed, as the temporal structure was clear without smoothing of the data. In all three cases Gaussian models were used to describe the temporal correlation, data for the theoretical models applied to the experimental semi-variograms being contained in the captions to the relevant figures.

The semi-variograms indicate that ^{99}Tc levels are relatively homogeneous from month to month and for separatory periods up to approximately 1 year but tend to vary substantially with separatory periods on the yearly scale. All three series exhibit evidence of trend in the data at the longer lag periods. This trend manifests itself within the semi-variograms as the rising portion of the experimental semi-variogram after lag periods of about 1 year. This trend is easily explained as within any one-year period ^{99}Tc values display little variation but from year to year levels have either increased (as for Hillesøy) or decreased (as for Utsira). The trend for the two Hillesøy series appears to occur at shorter lag periods than for the Utsira series and this may be caused by the Utsira being less sensitive (by virtue of location or oceanographic considerations) to processes causing the trend on what appears to be a yearly scale for the Hillesøy sets. As the trend does not manifest itself in any of the series before the sill value is reached, it plays little role in local estimation of values (i.e. estimation of values separated by less than the range of correlation from the weighted points). Due to this fact no attempt was made to detrend the data using the appropriate linear function.

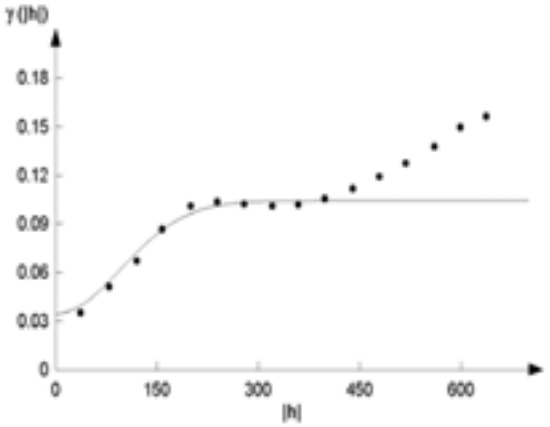

Figure 10. Semi-variogram for ^{99}Tc values in seawater (Hillesøy). Lag (x) axis – days, γ(h) axis – $(Bq/m^3)^2$. Model parameters – Gaussian, range - 235 days, sill – 0.081, nugget – 0.034.

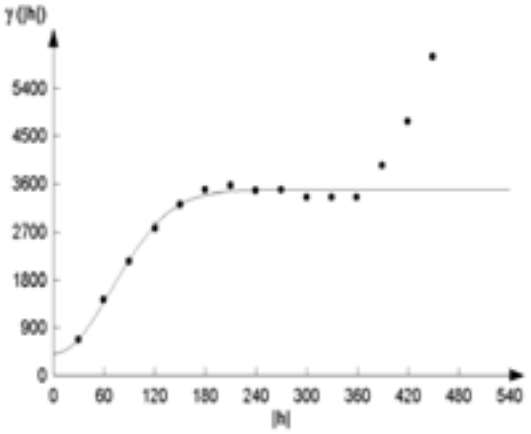

Figure 11. Semi-variogram for ^{99}Tc values in seaweed (Hillesøy). Lag (x) axis – days, γ(h) axis – $(Bq/kg)^2$. Model parameters – Gaussian, range - 167 days, sill – 3075, nugget – 420.

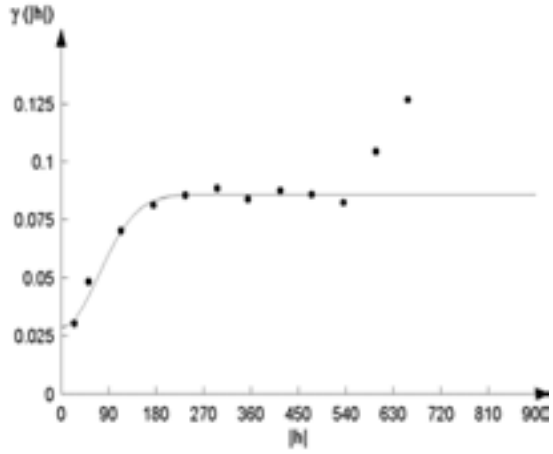

Figure 12. Semi-variogram for ^{99}Tc values in seawater (Utsira). Lag (x) axis – days, γ(h) axis – $(Bq/m^3)^2$. Model parameters - Gaussian, range – 174 days days, sill –0.057, nugget – 0.03.

3.2 Cross-validation

In order to ascertain the validity of the applied models with respect to the correlation range and to optimise some of the conditions for the subsequently applied estimation procedure, a cross validation exercise was conducted within GEO-EAS (Englund and Sparks, 1988). This involved sequentially eliminating one point from the data set, estimating its value from the remaining data using the temporal structure determined in the semi-variographic analysis and the estimation procedure, then reinserting the point and eliminating the next. The elimination/estimation procedure was repeated until all known points in the relevant data sets had been estimated by the chosen procedure. Comparison of the produced estimated values with the corresponding actual values and statistics related to the set of errors in the estimates allowed optimisation of the estimation process in relation to search radii and the numbers of known points required for estimation to be executed .

Cross-validation as a tool for determining appropriateness of applied models and estimation parameters is often used in geostatistical analysis but certain pertinent aspects of the procedure are often overlooked. Cross-validation techniques cannot determine the "quality of fit" of any model applied to the experimental semi-variograms as a cursory analysis of the kriging system shows that the sill parameter of the semi-variogram does not play a role in the assignation of kriging weights. Therefore the results of the analysis cannot be used to optimise the model in relation to the sill and by extension, the nugget value. Neither can cross-validation be applied reliably to sparse or unevenly spaced data sets. As the data in this study follows a semi-regular spacing pattern and the analysis was only used to adjust the range parameter and the search conditions for estimation, adoption of the procedure as described in the text above was deemed valid.

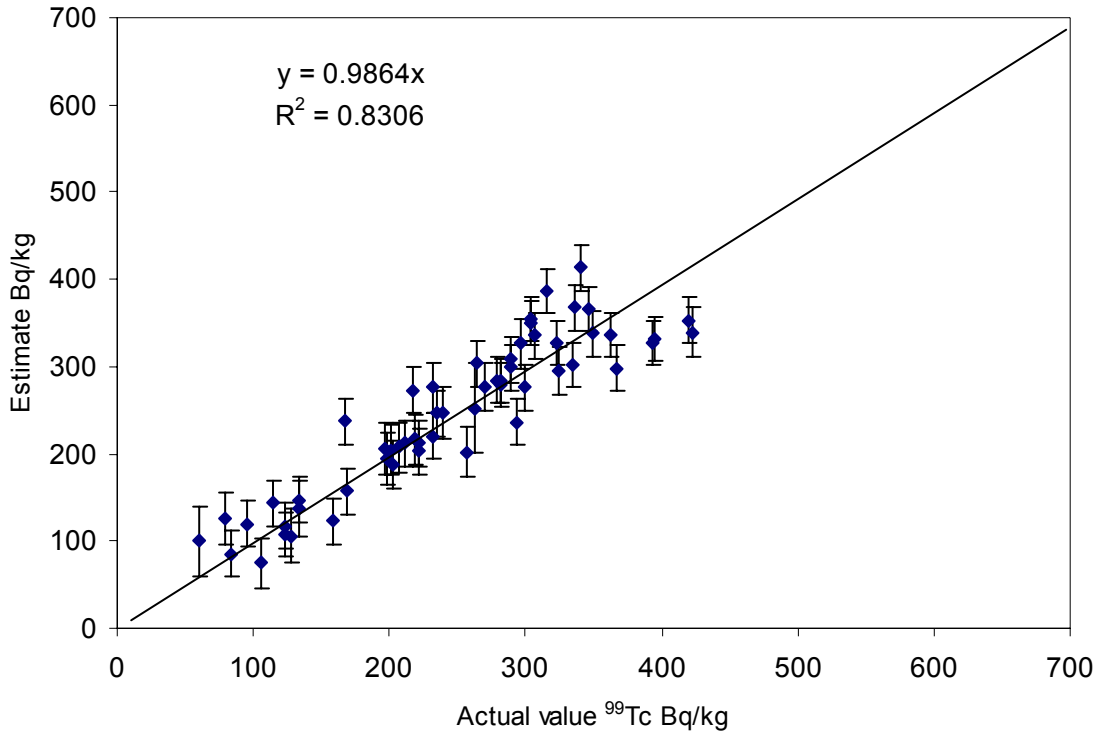

Figure 13. Cross-validation results for the seaweed series from Hillesøy.

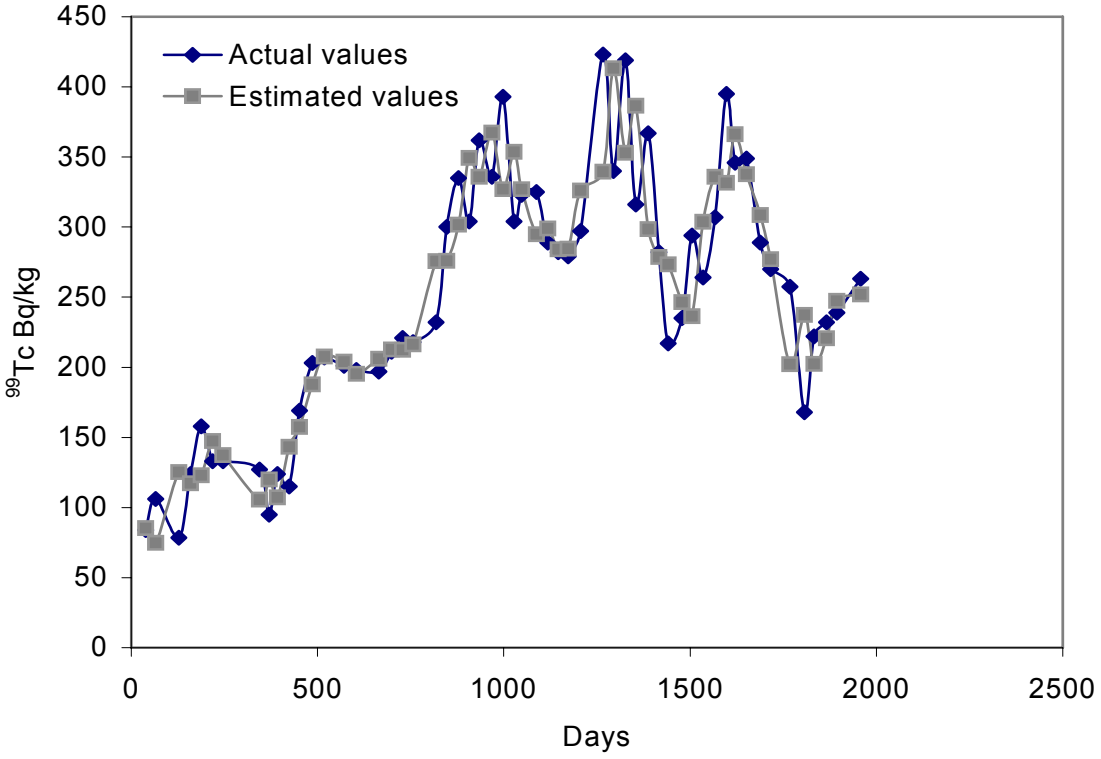

Figure 14. Reproduction of the seaweed time-series for Hillesøy using the cross-validation procedure.

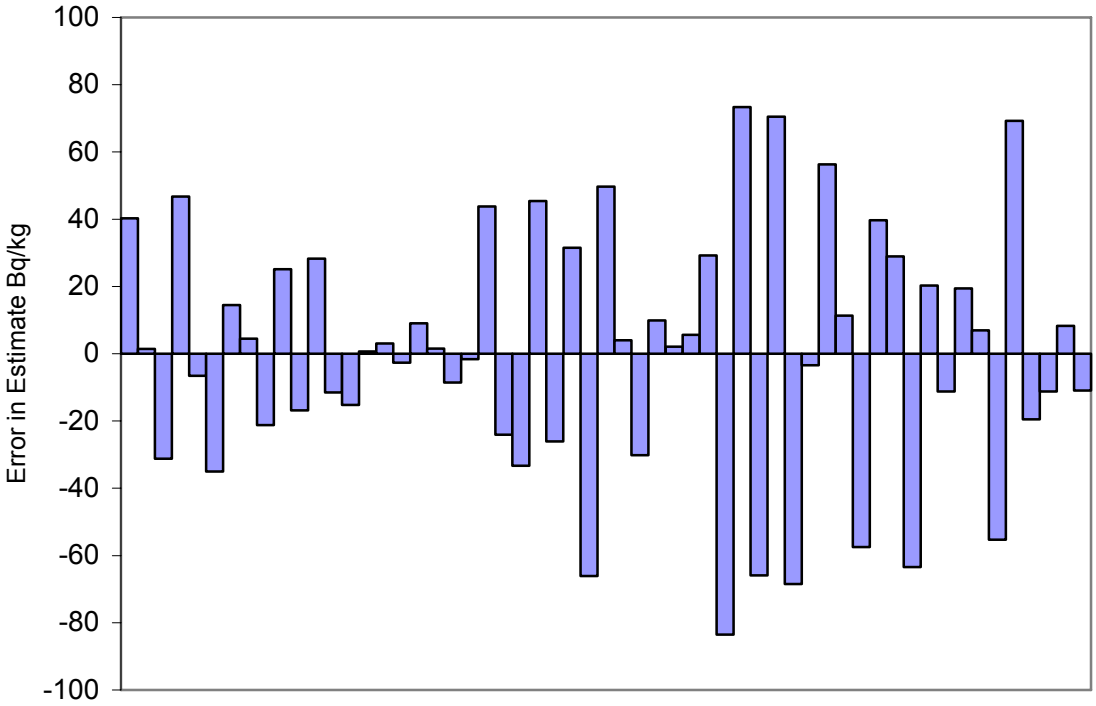

Figure 15. Distribution of over and under estimation errors for the cross-validation of the Hillesøy seaweed data set.

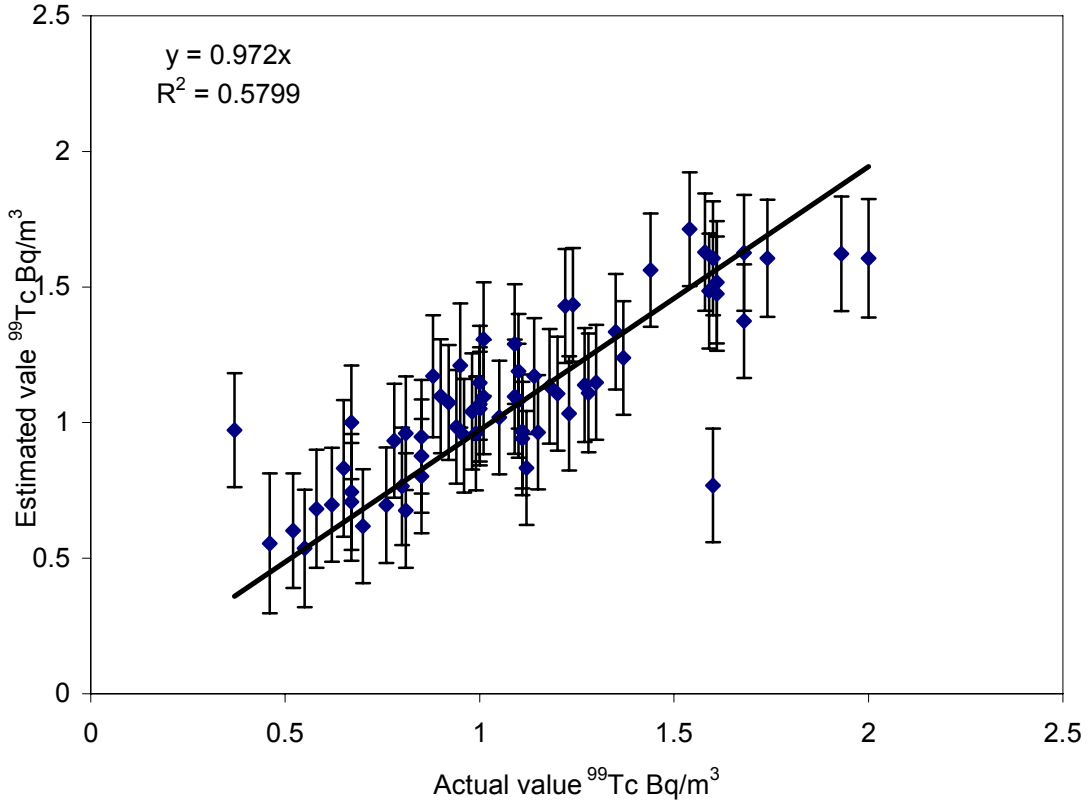

Figure 16. Cross-validation results for the seawater time series from Hillesøy

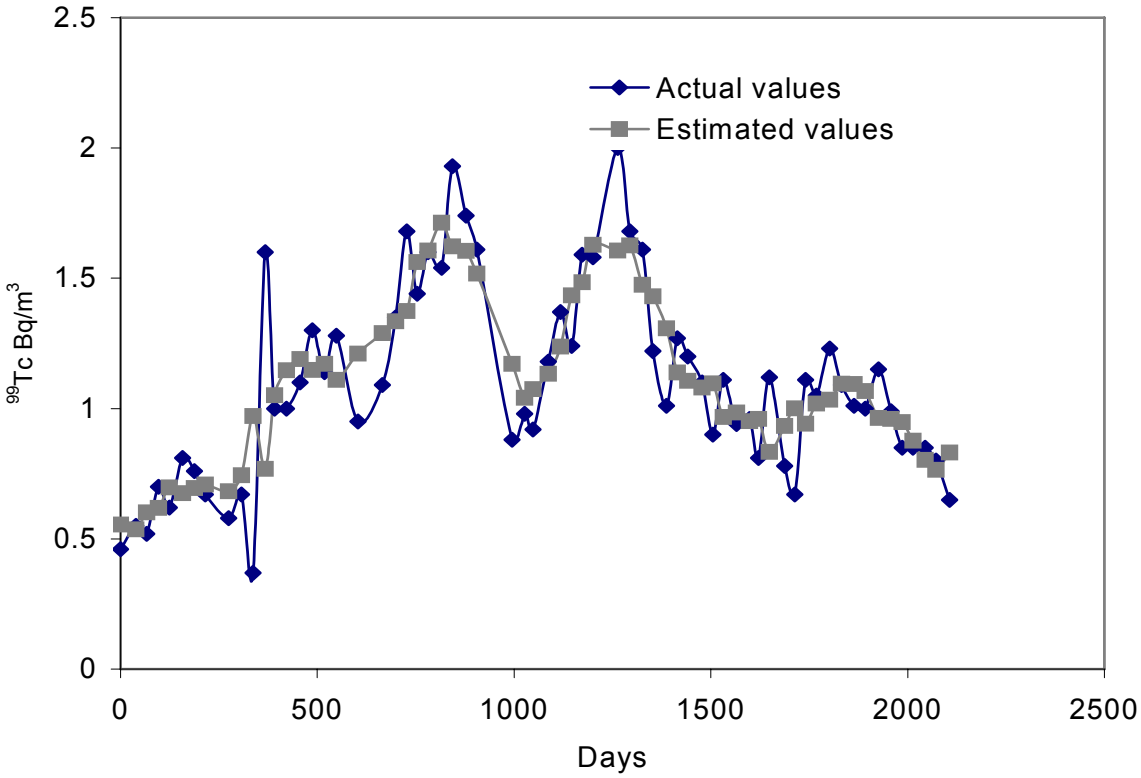

Figure 17. Reproduction of the seawater time-series for Hillesøy using the cross-validation procedure.

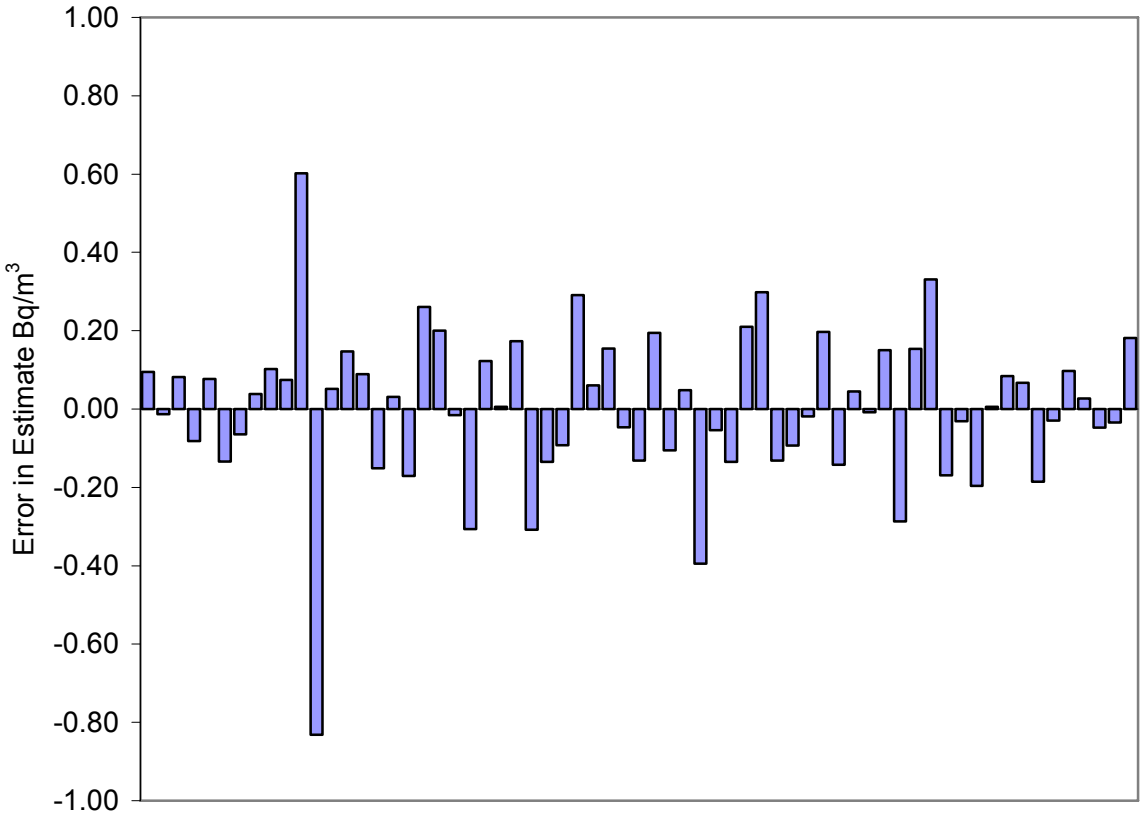

Figure 18. Distribution of over and under-estimation errors for the cross-validation of the Hillesøy seawater data set.

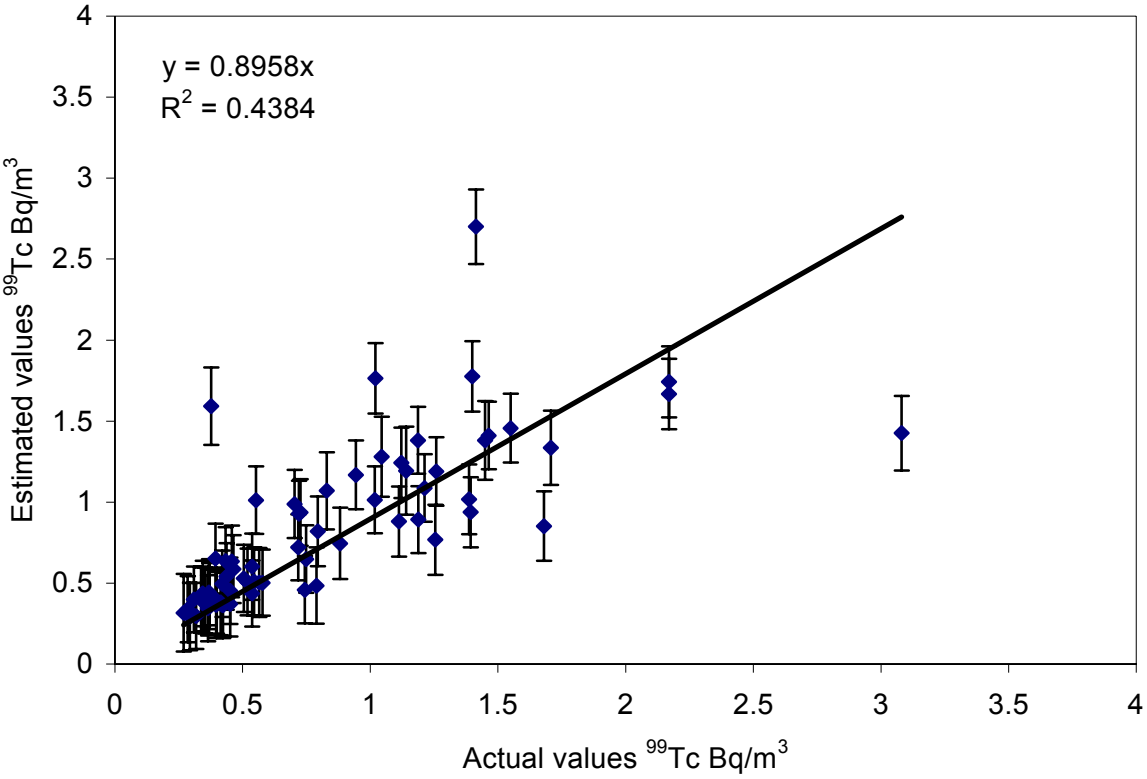

Figure 19. Cross-validation result for the seawater series from Utsira.

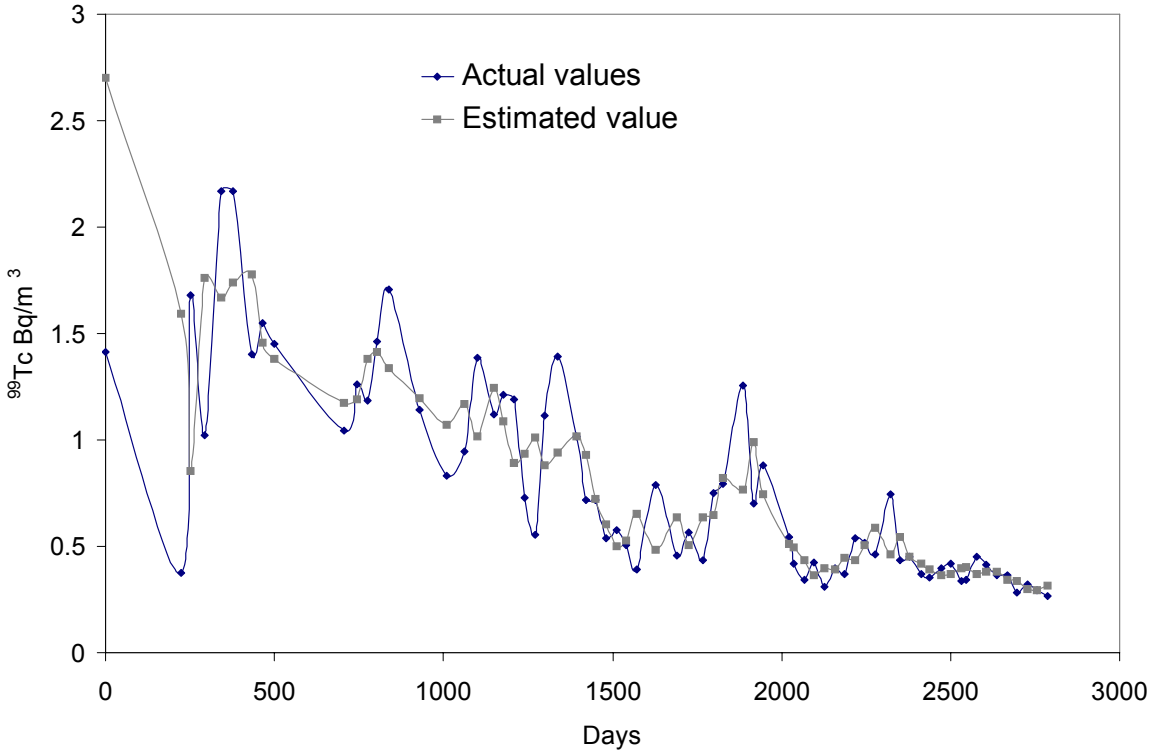

Figure 20. Reproduction of the seawater time-series for Utsira *using the cross-validation procedure.*

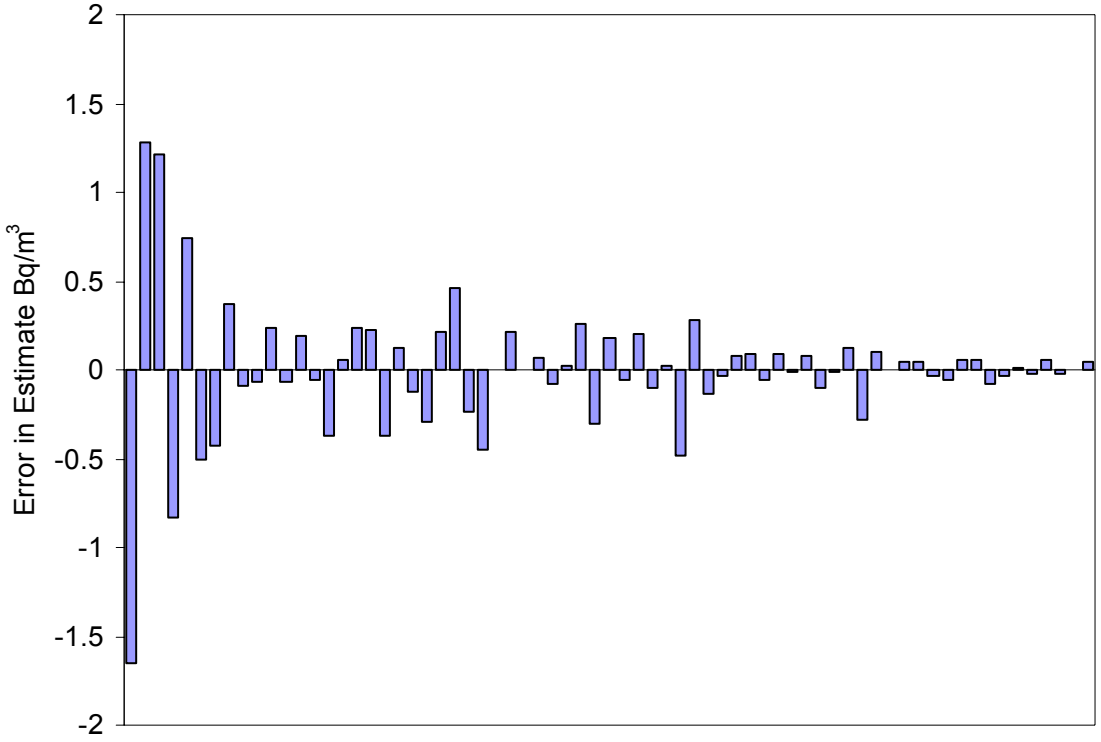

Figure 21. Distribution of over and under estimation errors for the cross-validation of the Utsira seawater data set.

As can be seen from the data (Fig.'s 13 - 21), the results of the analysis are variable while remaining acceptable. The best results were observed for the seaweed data set from Hillesøy. Strong correlation was observed between the actual and estimated values and the time series itself is accurately reproduced (Fig.'s 13 and 14). The distribution of the estimates indicates no global under or over estimation (Fig. 15). The accuracy of the estimates for the seaweed data can to some extent be explained by examination of the semi-variogram for this time-series. The nugget value, representative of the inherent randomness of the data, is relatively small compared to the sill value, which approaches the total semi-variance of the data. That this nugget value is small is most probably due to the fact that the analytical signal for seaweed is relatively high which reduces the associated uncertainty in the values (which contribute to the nugget parameter) and the data set itself is inherently smooth relative to the water series (due to the uptake mechanisms of the organism which smooth out the fluctuations in the corresponding water signal).

The lack of extreme values relative to the general data set means that the smoothing effect of the kriging process is less and the data is better reproduced at the extreme ends of the distribution, increasing the correlation between the actual and estimated values and resulting in a more even distribution of the estimation errors across the data. Results of the analysis for the Hillesøy seawater series are less impressive (Fig.'s 16 – 18) than for the seaweed series, some evidence of over and under-estimation being present although this would appear to be primarily limited to the two ends of the statistical data distribution. The majority of these points are for low and high values in close proximity to each other but the distribution of the errors remains even. The distribution of the time series estimates remains quite good. The uncertainties associated with the estimates are also proportionally higher than those associated with the seaweed data due to the greater contribution of the nugget component to the overall semi-variance of the data. The results for the Utsira data set (Fig.'s 19 - 21) are comparable to those for the Hillesøy seawater data although the extreme values encountered in this data set lead to isolated but

significant errors for some data points. These erroneous estimates are reflected in the reproduction of the distribution where it is clear that the earlier high values in the data are significantly underestimated. For this reason, an argument may be made for the elimination of what may be statistical outliers in the data but as no information was available as to ^{99}Tc levels prior to the start of this time-series the decision was made to remain as faithful to the empirical data as possible.

The cross-validation analysis serves to demonstrate that no significant bias exists in the estimation parameters employed and that realistic estimates can be obtained using the data to hand, the semi-variogram results and the estimation parameters employed in the study. The parameter of interest with respect to using such an estimation procedure for the optimisation of a sampling plan is the uncertainty in the estimates produced as part of the kriging procedure. This uncertainty is not the differrence between the actual and the estimate value but rather the uncertainty returned by the process known as the kriging standard deviation. Optimisation of sampling regimes with respect to sampling frequency must therefore be conducted using this parameter as the decision criteria.

4 Optimisation of Sampling: Kriging Error Minimisation

In order to demonstrate how minimisation of the kriging standard deviation could be employed as an aid to sampling regime design, it is worth considering the following scenario that although hypothetical will use the locations and data utilised in this study. The scenario suggested is that it is desired to obtain monthly values for ^{99}Tc in seaweed and water at Hillesøy and monthly values for ^{99}Tc in water from Utsira.

The temporal structures of the data sets are known from the actual samples. Information is required about how often samples must be taken at the locations in order to produce monthly values for ^{99}Tc in the relevant matrices for the purpose of monitoring trends in the levels of this isotope in the marine environment. Using the data to hand, the study simulates the effect of changes in sampling frequency on the quality of estimates produced. For example, using the Hillesøy data, the seawater time series covers a period of approximately 2100 days with samples having been taken every month approximately. In order to simulate a reduced sampling fre-quency, data was deleted until the chosen frequency was obtained. An attempt was made to keep the retained data as evenly distributed over the time period as possible. For each reduced data set, monthly values were estimated (every 30 days) and the uncertainty distribution was checked. Various parameters were then used to describe the effect of sampling frequency.

4.1 Hillesøy Seawater Data

In actuality, some 65 seawater samples had been taken over the 2100 day period at Hillesøy. This data was filtered to produce data sets with different numbers of samples as evenly spaced sample distributions and the monthly values were then estimated using the parameters described in the previous sections. Various statistics of the uncertainty distribution (the uncertainties as produced by the estimation) were then used to establish criteria for decisions regarding sampling frequency.

The impact of reduced seawater-sampling frequency at Hillesøy on the average kriging standard deviation of monthly ^{99}Tc estimates are displayed in Fig. 22. As can be seen, the uncertainty associated with the estimates is at its minimum for sample numbers approaching the actual number taken and begins to increase with reduced sample frequency. As can be seen from

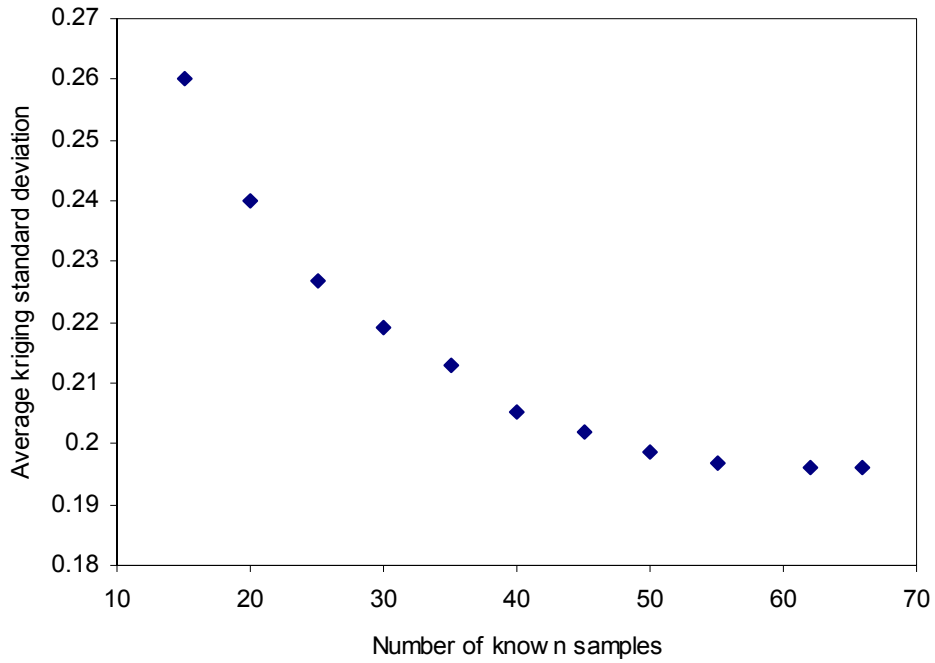

Figure 22. Variation in average kriging standard deviation as a function of sampling frequency for seawater from Hillesøy.

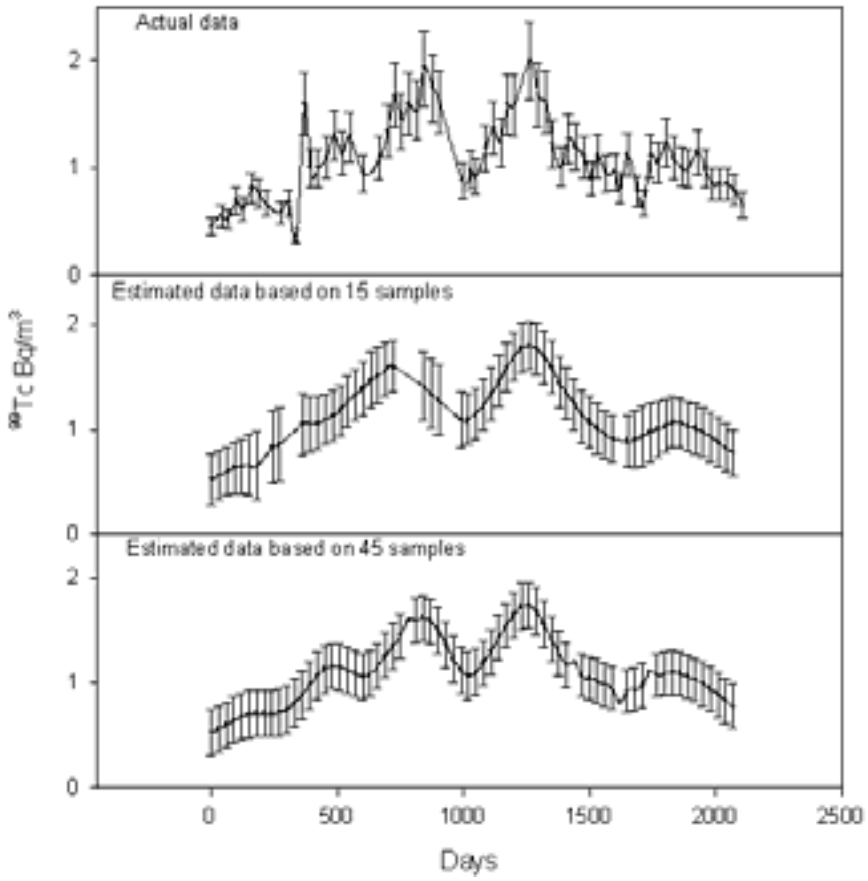

Figure 23. Representations of ^{99}Tc activity over time for seawater from Hillesøy based on the estimation procedure using 15 and 45 of the original data points compared with the actual data set. Error bars are the analytical uncertainty for the actual data and the kriging standard deviations for the estimates.

the graph however, the increase in uncertainty does not begin to rise dramatically until less than 30 samples were taken over the 2100 days, indicating that using the estimation procedure as described, monthly estimates of ^{99}Tc activity in seawater could be produced with half the number of samples taken, where the average uncertainty in the estimate would be comparable to the uncertainty in the data set resulting from 60 samples over the period.

As can be seen from Fig. 23, reduced sampling accompanied by an appropriate estimation of monthly values provides essentially the same information as provided by the more intensive "real" situation. With 15 samples taken over the period (sampling approximately every 4 months) some detail is lost, mostly where estimates could not be produced due to no known points lying within the kriging search radius (in which samples for local estimation must occur). The series produced by sampling every 6 weeks is substantially better, no increases in activity being missed by the produced series relative to the actual data. In many cases it can be seen that the uncertainty in the estimate is slightly larger than the uncertainty reported for the actual data but this difference is not extreme in any case. The smoothing effect of the kriging procedure is clearly evident in the data although even quite small features of the actual data are reproduced in the estimated data set.

Alternatively, the maximum kriging standard deviation could be used in a situation where information was required that could not have an uncertainty of greater than a certain value. Figure 24 displays variations in the maximum kriging uncertainty as a function of sampling frequency. The ascent is much sharper in this curve although the results indicate that little is to be gained for sample numbers greater than 40.

4.2 Hillesøy Seaweed Data

57 seaweed samples had been taken over a 1960 day period at Hillesøy. This data was filtered as for the seawater data from the same location to produce different evenly spaced sample distributions and the monthly values were then estimated. Figure 25 displays the effect of sampling frequency on the quality of the estimates produced by the kriging procedure. The results for the seaweed data set indicate that for the 1960 day period utilised, there is little to be gained with respect to estimate quality for sample numbers greater than approx. 50 (sampling every 40 days). Figure 26 reproduces the actual data series for a total of 20 samples and 40 samples over the period. As for the seawater data, it can be seen that the essential features of the actual series are reproduced using 40 samples but significant degradation is observed for the series that only uses 20 samples.

4.3 Utsira Seawater Data

71 seawater samples had been taken over a 2700 day period at Utsira. This data was filtered as for the seawater data from the same location to produce different evenly spaced sample distributions and the monthly values were then estimated. Figure 27 displays the effect of sampling frequency on the quality of the estimates produced by the kriging procedure. The analysis for the Utsira data indicates a marked cut-off in the uncertainty of the estimates at the 30 sample mark.

Reproduction of the time series based on a reduced number of samples (Fig. 28) is not as good as for the Hillesøy sets, less detail being displayed with respect to short scale variations in the levels of ^{99}Tc.

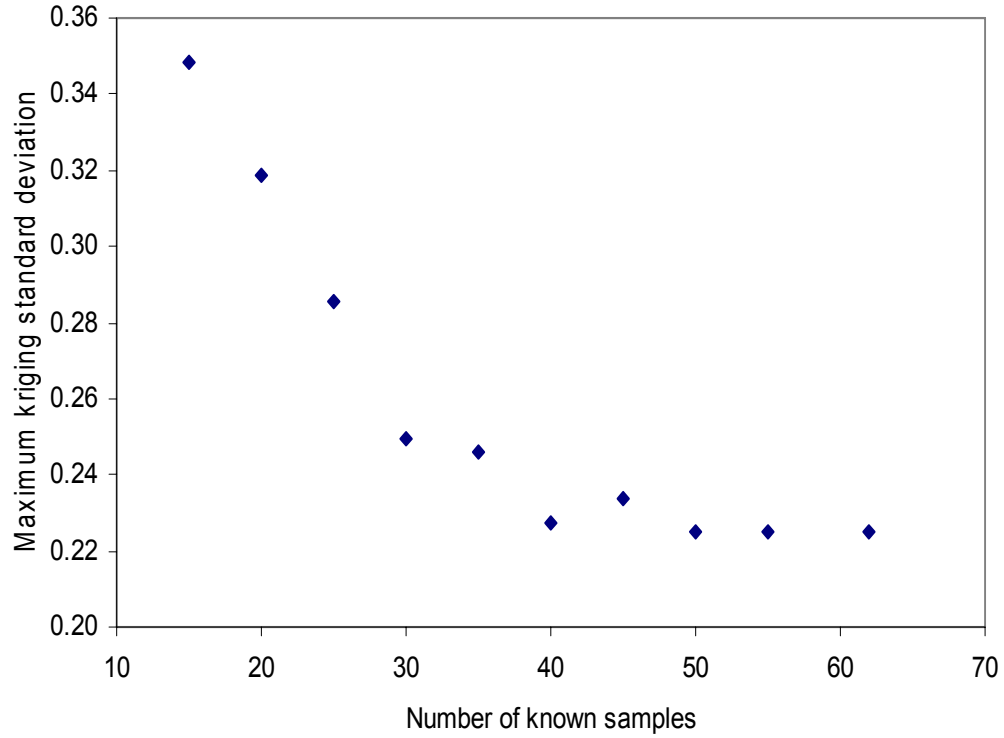

Figure 24. Variation in maximum kriging standard deviation as a function of sampling frequency for seawater from Hillesøy.

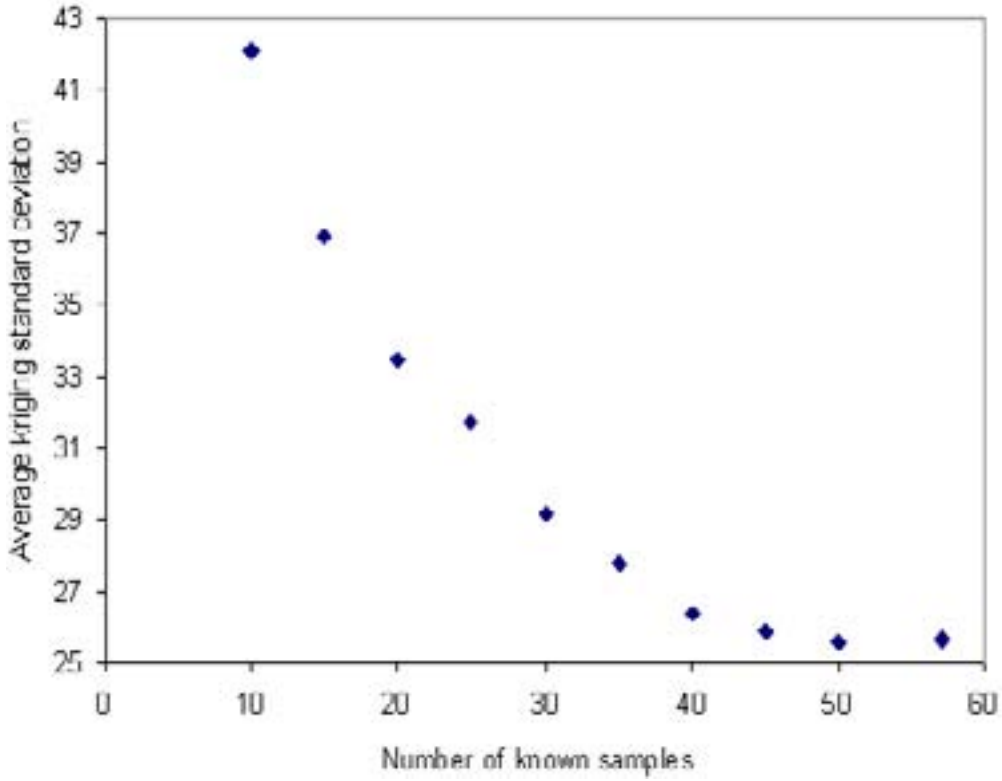

Figure 25. Variation in average kriging standard deviation as a function of sampling frequency for seaweed from Hillesøy.

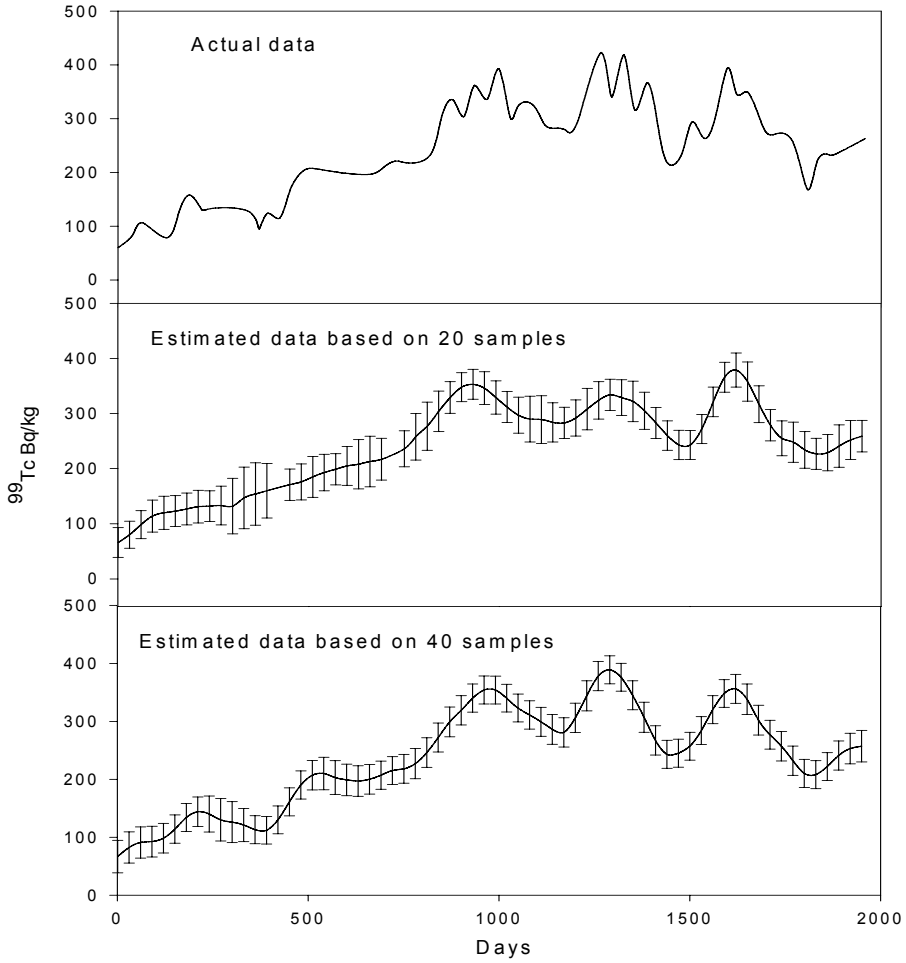

Figure 26. Representations of ^{99}Tc activity over time for seaweed from Hillesøy based on the estimation procedure using 20 and 40 of the original data points compared with the actual data set. Error bars are the kriging standard deviations for the estimates. Note: uncertainty data for actual series not provided.

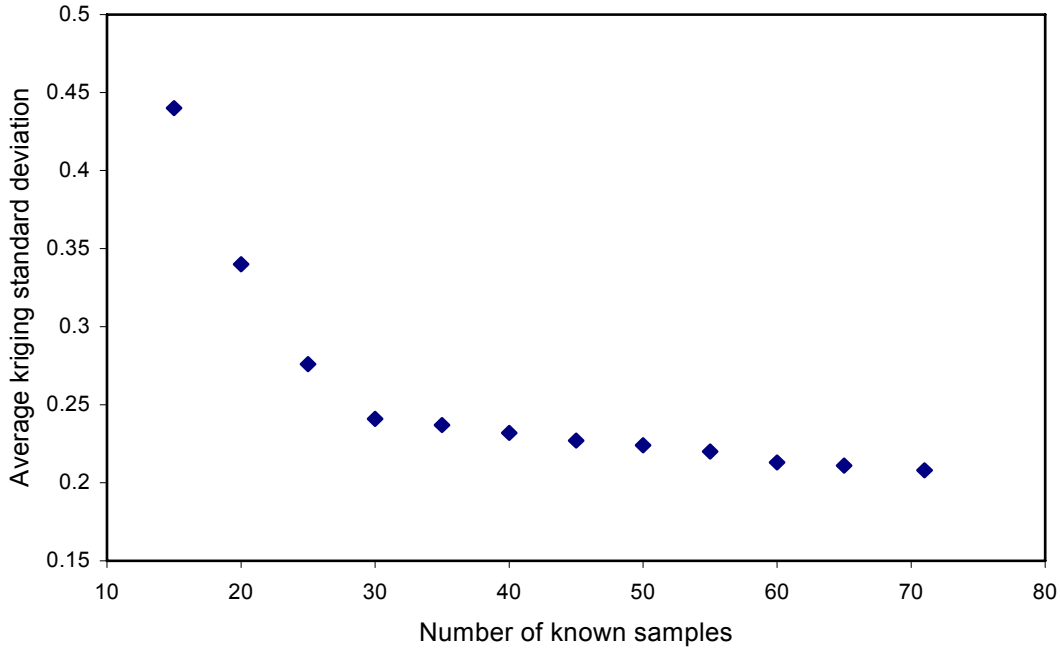

Figure 27. Variation in average kriging standard deviation as a function of sampling frequency for seawater from Utsira.

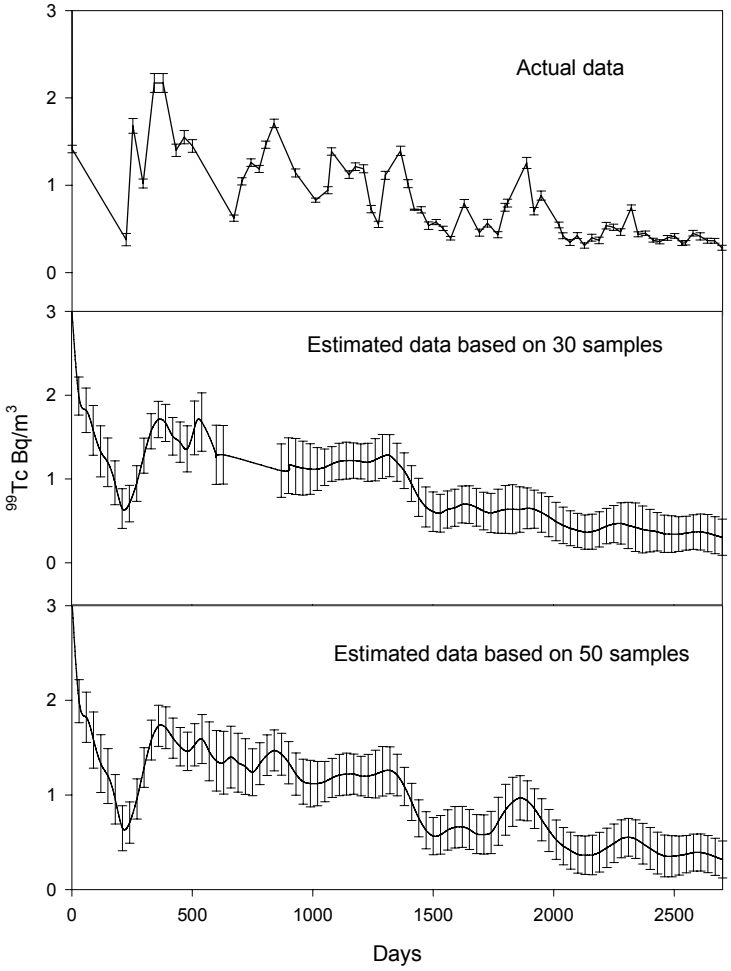

Figure 28. Representations of ^{99}Tc activity over time for seawater from Utsira based on the estimation procedure using 30 and 50 of the original data points compared with the actual data set. Error bars are the analytical uncertainty for the actual data and the kriging standard deviations for the estimates.

4.4 Analysis of Empirical Data Relative to Oceanographic Model Data

The above analyses, while providing an indication of how sampling frequency can affect the quality of estimates produced by the kriging procedure is inherently flawed for a number of reasons. None of the data sets are absolutely evenly distributed over the time periods involved therefore the selection of samples to be included in the reduced data sets may affect the outcome. Secondly, kriging is an "exact" interpolator in that if a point to be estimated coincides with a known point, the known data is honoured and returned as the estimate. Finally, due to uneven data spacing some points could not be estimated especially for the much-reduced sets. These points were omitted from the statistics and may affect the overall outcome.

The above discussion has demonstrated that, for the given example of producing monthly values for ^{99}Tc (or, theoretically, any long-lived radionuclide), it is possible to produce a series of monthly data with associated uncertainty using a set of samples taken on less than a monthly basis provided that:

- a level of temporal correlation exists for the data,
- the semi-variogram has been calculated and correctly modelled and that the

data conforms to certain constraints (regarding the intrinsic hypothesis),

- the kriging estimation procedure has been optimised.

Determination of how many samples should be taken can then be based on the desired level of uncertainty in the estimated data points and the frequency for which estimated points are to be produced. The problem remains however that knowledge of the semi-variogram structure must be obtained in some way prior to the optimisation of the sampling. In the design of monitoring schemes for, for example, determination of contaminant levels in soil, this is not such a problem as a one off exploratory sampling campaign can be performed prior to the design of the monitoring scheme. With temporal monitoring this is more difficult as acquiring a data set for semi-variography may necessitate years of sampling. It is possible that literature data may be used or that time series from other sites may be used although both of these approaches require a high level of presumption regarding the extrapolation of data from one site to another. Assuming that the temporal correlation exhibited by a data set is to some extent a function of the processes involved in the occurrence of the contaminant at the location being studied, it may be possible to calculate the semi-variogram from modelled data without relying on the prior existence of a large data set. To test this approach, use was made of modelled ^{99}Tc data. The North Atlantic-Arctic Ocean Sea Ice Model (NAOSIM) (Gerdes et al., 2001; Karcher et al., 2003, 2004), developed at the Alfred Wegener Institute for Polar and Marine Research, is a 3D hydrodynamic coupled ice-ocean model, which covers the Arctic Ocean, the Nordic Seas and the North Atlantic north of 50° N. The model is driven by daily atmospheric data from the NCEP/NCAR reanalysis dataset covering the period from 1948 to 2002. The NAOSIM model run used for the present study features a 10 m vertical division of the upper ocean (33 levels in total). Hydrographic initial

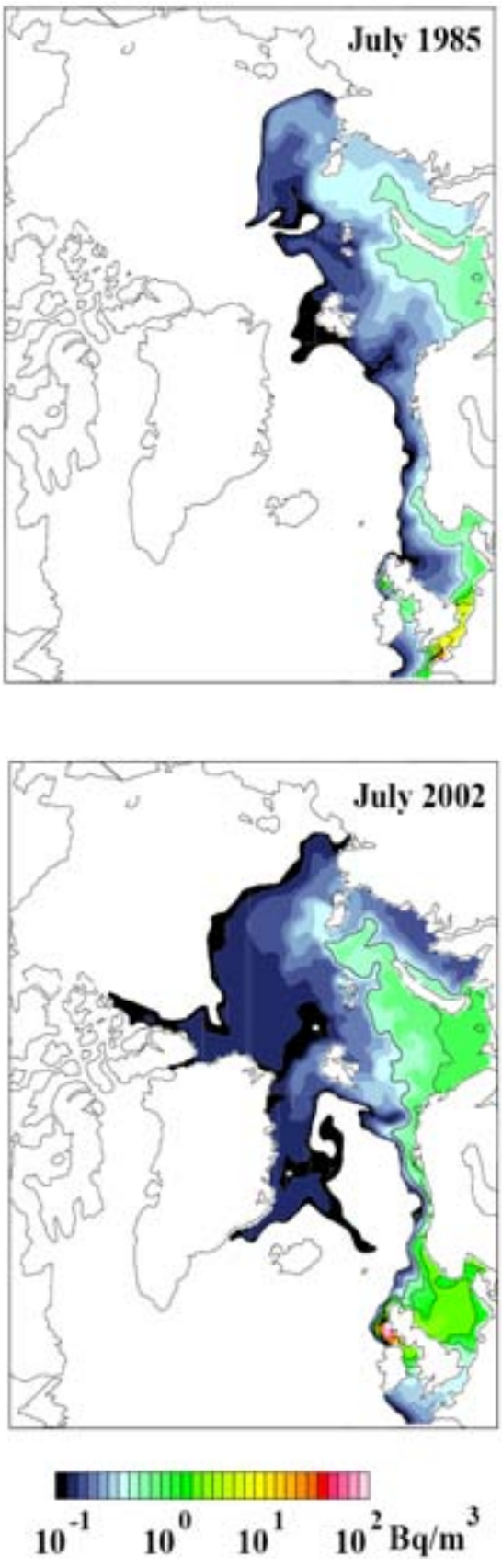

Figure 29. Simulated dispersion and transport of ^{99}Tc along the Norwegian coast for the periods July 1985 and July 2002.

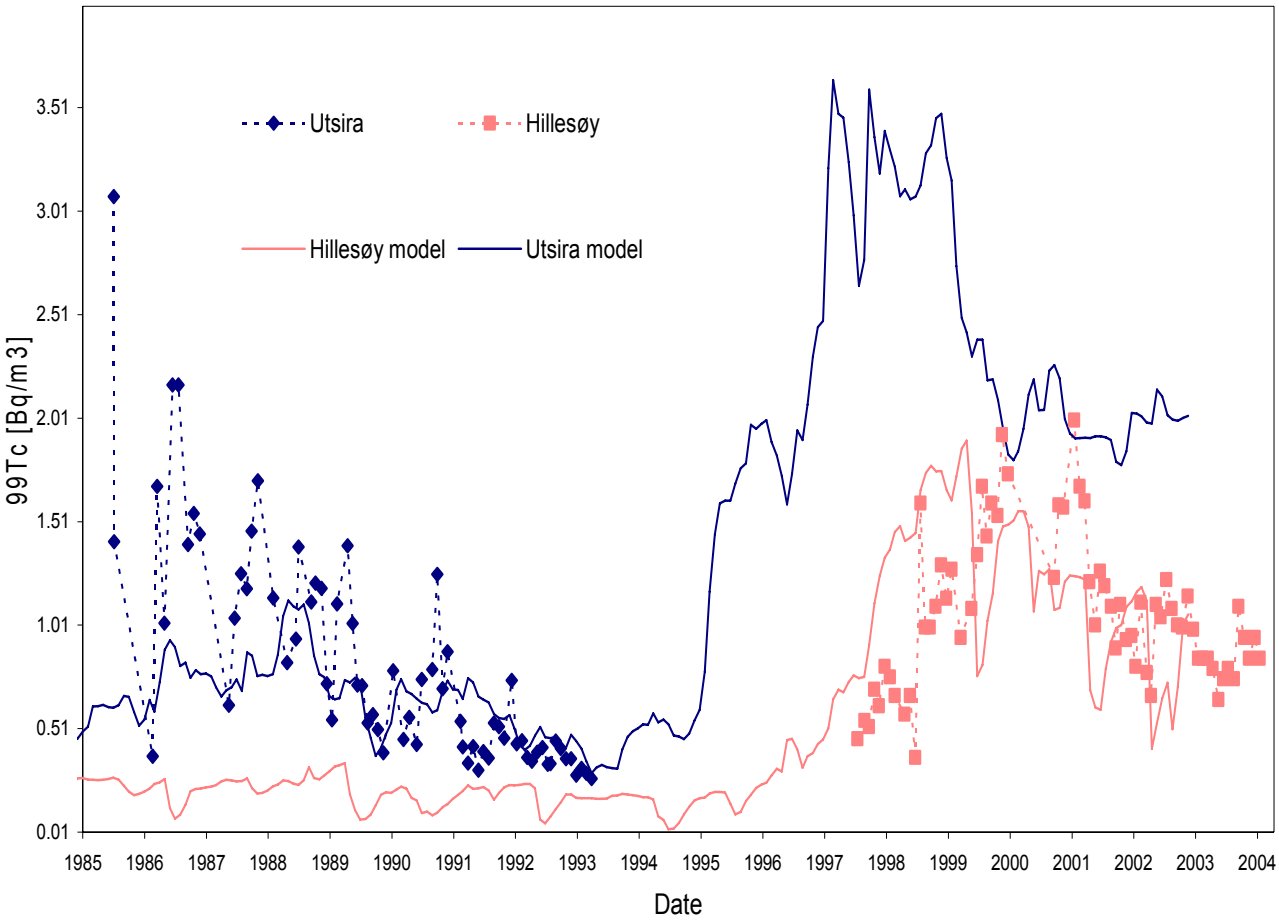

Figure 30. Modelled and actual data for Hillesøy and Utsira ^{99}Tc values.

conditions for the latest model runs are based on the most recent version of the Polar Science Centre Hydrographic Climatology (PHC), while run-off from major rivers into the model domain, including diffuse run-off and run-off from the Norwegian coast, are included. The model resolution is approximately 28 km. After 1970, the model is subjected to an input of ^{99}Tc near the location of the nuclear reprocessing facilities in Sellafield. Simulated results from the model for the dispersion and transport of ^{99}Tc along the Norwegian coast for two time points of relevance to this study are depicted in Figure 29. While the ^{99}Tc release data are provided as yearly means from 1970 to 1994, monthly mean data are provided for the period thereafter. Simulated concentrations at Hillesøy between the beginning of 1971 and the end of 2002 are depicted in Figure 30.

As this model incorporates the hydrodynamic processes that govern the occurrence of ^{99}Tc in seawater at Hillesøy and Utsira, it was decided to ascertain to what extent the model data replicated the temporal structure as exhibited by the actual data values. To achieve this, the modelled data sets were subjected to semi-variographic analysis as for the actual data. For the Hillesøy data set the semi-variograms were established over the time period for which actual data was available this corresponds to the period between July 1997 and May 2002 as this is the extent to which the model data has been calculated. The calculated semi-variograms and fitted models are displayed in Figure's 31 and 32.

For both sets of data, the semi-variograms are comparable with respect to the range of correlation and the sill although nugget parameters exhibit some difference between modelled and actual data.

The nugget values for the semi-variograms produced for the model data are closer to zero that for the actual data. As the nugget is representative of the unstructured or random component to the overall semi-variance (i.e. samples taken at exactly the same time will have slightly different values), it is likely that analytical error, sampling error and the other uncertainties with respect to the analytical procedure contribute largely to this value. The modelled data on the other hand does not include such uncertainties and is therefore more likely to produce a lower random contribution. The range however, which is the parameter of most interest in the design of the sampling plan, for the semi-variogram of modelled data, is quite comparable to that produced by the actual series. The comparability of the semi-variograms produced may indicate that in the case of designing a sampling scheme at a location where no previous data exists, it may be possible to employ models that describe the processes affecting the temporal structure of the data to make an a priori assessment of the semi-variogram.

Furthermore, the model provides the possibility to investigate whether a monitoring regime is capable of reflecting certain hypothetical discharge or accidemt scenarios by using different discharge datasets in the model run.

In order to observe the temporal structure as exhibited by the modelled Hillesøy data over long range time-scales (years instead of months), a semi-variogram was constructed covering the entire range of the models data. This is presented in Figure 33.

As can be seen from Fig. 33, long range modelled data indicates that ^{99}Tc values in seawater cannot be considered to be random up to separatory periods of approximately two years and a second long term correlation begins to come into effect after approximately six years. Such long range correlation does not negate the validity of the short term structure exhibited by the short range semi-variograms but appears to indicate phases to the temporal structure of ^{99}Tc concentrations in seawater at Hillesøy.

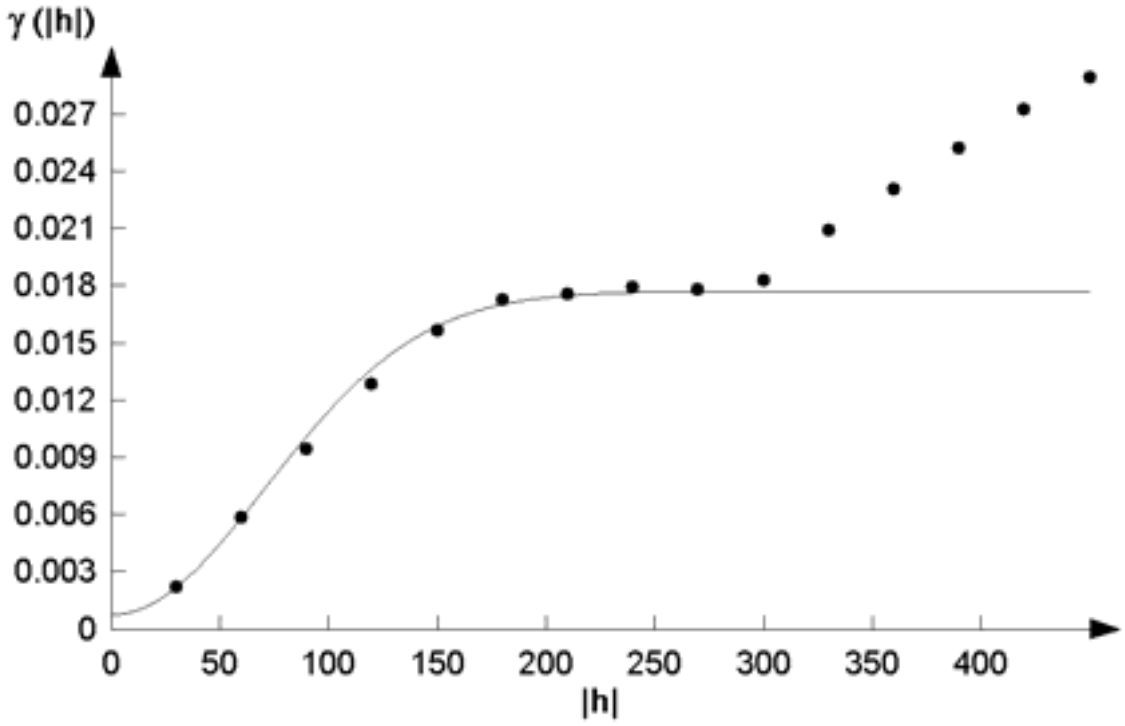

Figure 31. Semi-variogram for modelled ^{99}Tc values in seawater (Utsira) over the time scale of the actual data. Lag (x) axis – days, γ(h) axis – (Bq/m³)². Model parameters – Gaussian, range - 178 days, sill – 0.017, nugget – 0.0008.

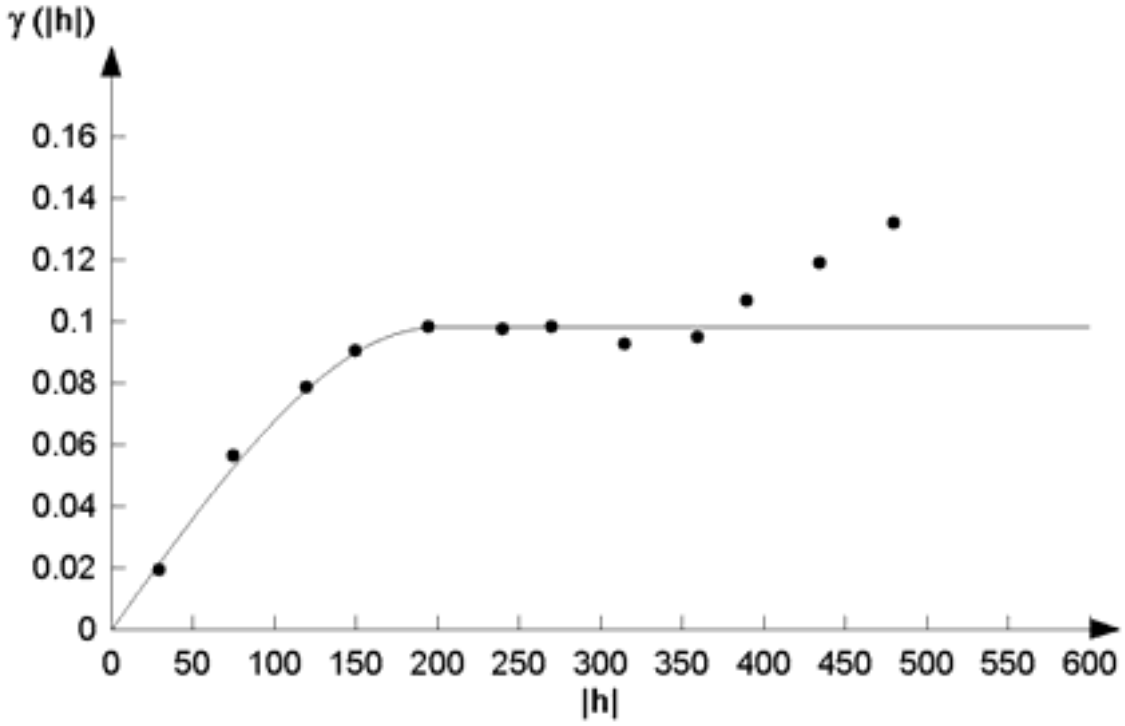

Figure 32. Semi-variogram for modelled ^{99}Tc values in seawater (Hillesøy) over the time scale of the actual data. Lag (x) axis – days, γ(h) axis – (Bq/m³)². Model parameters – Spherical, range - 210 days, sill – 0.095, nugget – 0.0.

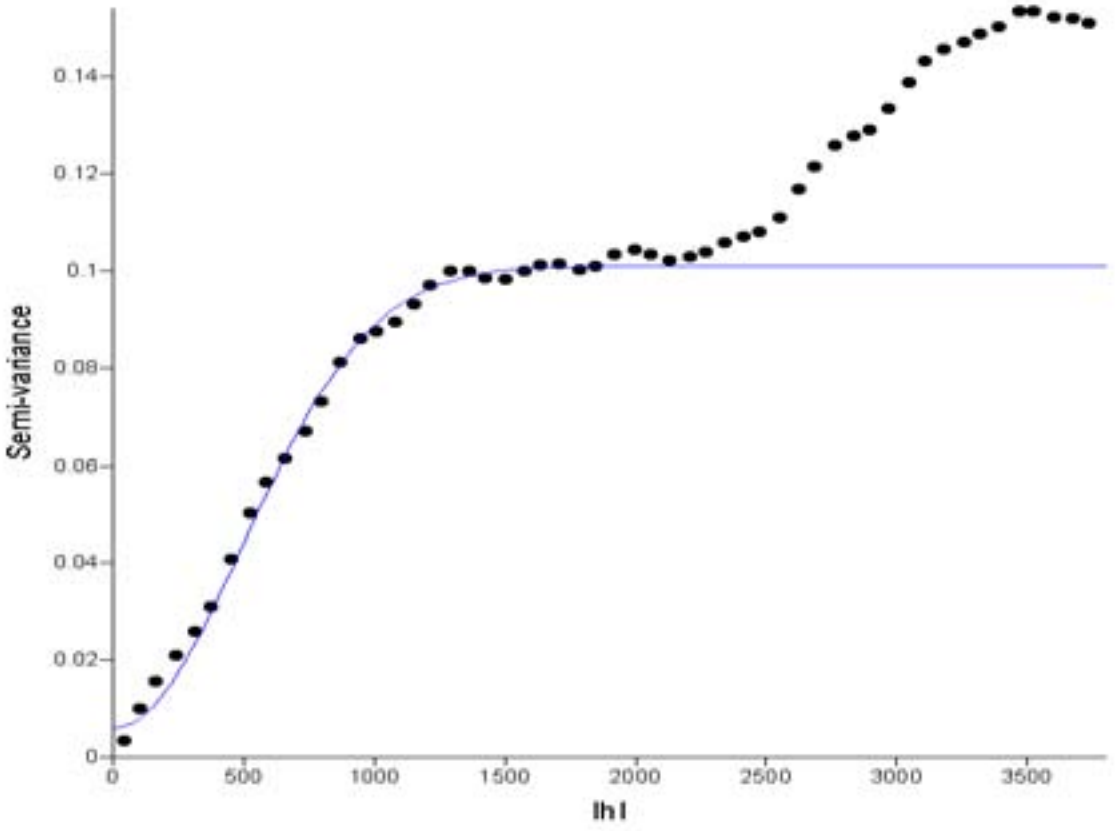

Figure 33. Semi-variogram for modelled ^{99}Tc values in seawater at Hillesøy. Lag (x) axis – days, γ(h) axis – (Bq/m³)². Model parameters – Gaussian, range - 700 days, sill – 0.1, nugget – 0.0008.

Trend or non-stationarity is evident in the data as the sills do not remain constant; the sill is attained after separatory periods of some months then the semi-variance rises again to a sill for periods of two to three years and the final apparent sill is attained for periods of approximately ten years. It is not known what processes govern the various stages exhibited by the long range semi-variogram although it is probable that there may dominant processes for each step. A hypothetical example could be that seasonal factors are contributing to the behaviour at the monthly scale, perhaps oceanographic considerations on the tri-yearly scale and changes in the source terms on the scale of decades.

5 Discussion

For the monitoring of concentrations of ^{99}Tc (or any other radionuclide or contaminant) in the marine environment, there are essentially four approaches that may be adopted in determining the number of samples to be taken to achieve the desired objectives; in this case obtaining an overview of trends in concentrations of the contaminant species in the marine environment. Random sampling makes no assumptions about the distribution or movement of the analyte species and tends to involve the taking of more samples than other sampling approaches. Systematic sampling also makes no assumptions regarding the factors introduced above. Judgemental sampling regimes do make assumptions regarding movement and distribution with time or space. Combination sampling is the implementation of a sampling regime based on the use of any combination of the previous three types of regime. The data used in this study can be described as a combination of judgemental and semi-systematic sampling given that some information about the processes governing the distribution of the analyte were known prior to the initiation of the regime and the sampling is conducted on an approximately systematic basis.

The goal of the current sampling regime is to describe the variation in ^{99}Tc concentrations in the marine environment with time. As discrete samples are taken, some form of interpolation is necessary to describe the concentration of the contaminant at points intermediate between the discrete sampling points. The interpolator used is typically a straight line relationship between discrete points. This study adopts the approach that the quality of the results produced by the interpolation can be used as the foundation for determining the optimum sampling frequency. In planning any sampling regime, it is advantageous that the sampling frequency should involve minimum estimation or interpolation error for periods during which samples were not taken or minimum sampling effort for a given accuracy. Two approaches for matching sampling frequency to estimation or interpolation accuracy may be used: a classical approach, which ignores the temporal dependence between observations, and utilises classical "random" statistics (in this case Power Analysis) and a geostatistical approach, which exploits, in this instance, the temporal dependence displayed by the variable in question. To demonstrate the use of a geostatistical procedure, the temporal variability of ^{99}Tc concentrations in seawater and seaweed from two different locations was determined and this information was then used to determine how well different sampling regimes could reconstruct monthly values, by interpolation, for the concentration of ^{99}Tc in the sampled matrices. The criteria used to determine the efficacy of the sampling regimes was the average kriging error, the sampling regime producing the minimum error being deemed the most effective.

The results of the study indicate that that using a geostatistical estimation approach towards the production of monthly estimates of ^{99}Tc concentrations, there is little to be gained (with respect to the uncertainty associated with the estimates for unsampled times) by sampling at frequencies greater than approximately every 40 to 60 days depending on the matrix and

location involved. Some difference was observed between sites with respect to optimum sampling frequency and this could be related to how the sites physical location affects the temporal structure of the data.

In relation to the actual sampling regime currently employed, it would appear that, taking into consideration the practicalities of such sampling, that the current approach (sampling approximately every 30 days) provides a relatively effective regime for the monitoring of the isotopes concentrations, in relation to the information provided for the effort expended in the sampling and analysis. The results of the analysis indicate that more frequent sampling would not significantly improve the quality of information that may be provided by an implemented estimation procedure in relation to concentrations of the isotope for unsampled dates. The frequency of sampling as it currently stands also provides a useful level of leeway in that periods where samples may be missed (due to weather, sea conditions etc.) would be unlikely to significantly impact the overall quality of estimates produced using the remaining samples. The results of the analysis provide a useful rationale for the currently implemented regime in that the sampling frequency employed can be defended on the basis of a scientific analysis.

The use of the oceanographic model as the basis for the design of a sampling regime based on, in this case, geostatistical procedures, appears to offer certain distinct advantages. The first and most obvious of these would be the fact that such a model can provide information prior to any sampling having been conducted. It is of course accepted that any model used for such purposes must have had an acceptable level of validation performed to ensure its applicability but such validation would appear to be only necessary for one location. If that location does not differ significantly from an oceanographic standpoint from a second location, then it would appear that there is no reason such a model could not be used to design sampling regimes for the second location. For both locations used in this study, the model reproduced the temporal structure of the data acceptably with respect to the semi-variograms parameters of most importance to the procedures implemented. The ability to devise sampling regimes on an *a priori* basis confers significant advantages with respect to designing monitoring programs. It would appear that it may be possible to establish, in the early phases after a hypothetical release, the optimum sampling frequency required to determine with a given confidence level when a contaminant signal has reached a particular point. This could possibly be achieved by inputting the release scenario parameters to such a model, running the model to simulate a period of time sufficient to determine the temporal structure and then devising a sampling regime that is sufficient to reduce the interpolation uncertainty to a satisfactory level.

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