# Spectral and geostatistical analysis of measured and modelled Technetium-99 timeseries data in the Nordic marine environment

S. Gerland<sup>1</sup>, M.J. Karcher<sup>2,3</sup>, M.Dowdall<sup>4</sup>, D. Divine<sup>5</sup>, M. Iosjpe<sup>6</sup>, V. Pavlov<sup>1</sup> & J.P. Gwynn<sup>4</sup>

#### INTRODUCTION

Radioactivity in the Arctic environment is a central topic within environmental pollution issues (e.g. AMAP 1998). However, the way radionuclides spread with ocean currents is also interesting from an oceanographic point of view. Radionuclides discharged to the sea represent tracers which give the possibility to quantify long-distance transport of water masses both in spatial and temporal dimensions. Within the last decades, increased activities were initiated in order to monitor and model the occurrence of radionuclides from discharges into the North Atlantic and Arctic, including determination of transit times (e.g. Brown et al. 2002; Kershaw & Baxter, 1995; Kershaw et al. 1999). One of those, the radionuclide Technetium-99 (Tc-99, halflife 213 000 y), is a man-made, highly soluble, a beta emitter, that has been discharged from European nuclear reprocessing plants at Sellafield (U.K.) and La Hague (France). Tc-99 observed in 1988-90 in the East Greenland Current is combined of 85% originating from European coastal discharges and 15% from nuclear weapon test fallout (Dahlgaard 1994). Technetium-99 is transported to the high Arctic via the Norwegian Coastal Current and the West Spitsbergen Current, a branch of the North Atlantic Current (Gerland et al. 2003; Karcher et al. 2004), and via another branch, the North Cape Current, entering the Barents Sea. Since both sampling and laboratory analysis procedures are expensive and time consuming, radioecological monitoring is limited and needs to be optimized.

## SAMPLING AND MEASUREMENTS

Sampling for Tc-99 analysis for locations with relatively low concentrations requires samples of 50 l seawater. The surface water sampling was predominantly undertaken on a monthly basis from coastal locations in Northern Norway since 1997 (Hillesøy), Bjørnøya and Hopen since 2000 (both islands in the Barents Sea), Ny-Ålesund since 2000 (West-Spitsbergen) and the island Jan Mayen since 2001 (Greenland Sea). The setting of locations is strongly reflected in the signals observed, with relatively higher levels in the south, lower in the north and very low levels in the Arctic. Samples were analysed in the laboratories of the NRPA in Tromsø and Østerås, Norway, using a method involving ion-exchange chromatography and low level beta counting (Chen et al. 2001). Measurement errors are on average  $\pm$  0.05 Bq/m³.

<sup>&</sup>lt;sup>1</sup> Norwegian Polar Institute, Polar Environmental Centre, NO-9296 Tromsø, Norway, <sup>2</sup> Alfred Wegener Institute for Polar and Marine Research, P.O. Box 120161, 27515 Bremerhaven, Germany, <sup>3</sup> O.A.Sys - Ocean Atmosphere Systems, Schanzenstr. 36, 20357 Hamburg, Germany, <sup>4</sup> Norwegian Radiation Protection Authority, Environmental Unit, Polar Environmental Centre, 9296 Tromsø, Norway, <sup>5</sup> University of Tromsø, Department of Mathematics and Statistics, 9037 Tromsø, Norway, <sup>6</sup> Norwegian Radiation Protection Authority, P.O. Box 55, 1332 Østerås, Norway.

### DATA ANALYSIS AND MODELLING

As a part of the research project "RADNOR", time series from monitoring measurements and modeling of Technetium-99 activity concentrations for locations on the coast of southern and northern Norway and in the Arctic were analysed for standard statistical parameters. Hillesøy monitoring data (since 1997) and time series from NAOSIM modelling, covering the time period from 1971 to 2002, were additionally analysed with geostatistical methods (calculation of semi-variograms, Dowdall et al. 2003; Dowdall et al. in press), and the model results from NAOSIM also with a spectral analysis method (wavelet analysis, Percival and Walden 2000). Two models were used in this study. First, a hydrodynamic coupled ice-ocean model "NAOSIM" (Gerdes et al. 2001, Karcher et al. 2003), and second the NRPA compartment model (Iosjpe et al. 2002). Using the NAOSIM model, concentration data for the time 1971-2002 were calculated (see time series in Fig. 1). For the compartment model, experimental <sup>99</sup>Tc data sets for different marine regions (east, west and central parts of the Irish Sea, east and west parts of the Scottish Waters, central, north, south and east parts of the North Sea and Norwegian Sea) were used for model testing and evaluation (Fig. 2).

## **RESULTS AND CONCLUSIONS**

Measured <sup>99</sup>Tc data from five Arctic/Subarctic monitoring stations (Fig. 1) and from the Irish Sea, Scottish waters, The North Sea and Norwegian Sea (Fig. 2) show relatively good agreement with advanced oceanographic modelling. For the high Arctic sites, data are sparse, but mean levels are in the same order of magnitude as model results for those locations. Problems and time lags between modelled and measured signals can occur through local effects at sampling stations (e.g. freshwater run-off, tidal effects, bathymetry, currents and sea ice formation and melt), and to limitations in the models used.

The geostatistical analysis revealed that for a location like Hillesøy, a sampling interval shorter than approximately 50 days improves the quality of estimates for concentrations only little at a given time. With the combination of such analyses and advanced oceanographic modelling, monitoring strategies can be significantly supported and improved. However, here only surface water concentration measurements were considered. From NAOSIM model runs, existing knowledge of the ocean currents and *in situ* sampling (Gerland et al. 2003) one knows that substantial water mass transport (such as in the West Spitsbergen Current) can be dominated by currents located deeper than surface currents. For measurements of those concentrations, advanced sampling during scientific cruises is necessary.

Wavelet analysis show changes in wavelet coefficients for the Hillesøy NAOSIM time series especially for the early 1980s and late 1990s – periods when also the concentrations were increased relatively to the times before.

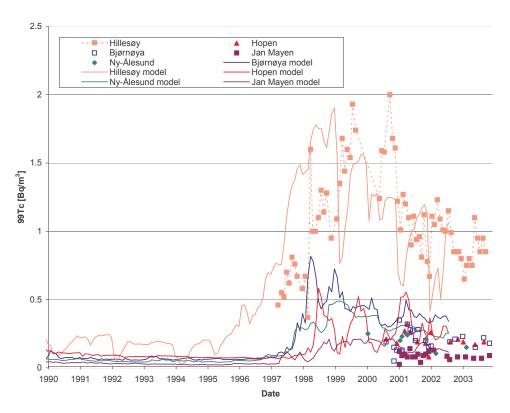
## **ACKNOWLEDGEMENTS**

We are grateful to A.K. Kolstad, B. Lind, Ø. Sælnes and C. Davids (all NRPA) for assisting with Tc-99 concentration laboratory measurements. We thank H. Ludvigsen (Hillesøy), personnel at the meteorological stations at Bjørnøya and Hopen (Norwegian Meteorological Institute), at the LORAN-C station Jan Mayen (FLO/IKT) and at the Sverdrup Station Ny-Ålesund (Norwegian

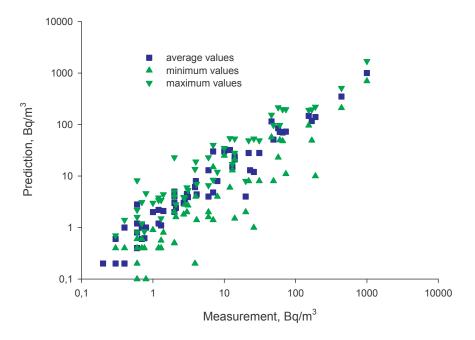
Polar Institute) for taking seawater samples. Data used for the NRPA compartment model evaluation origin from sampling during several cruises (Marine Monitoring Programme, "RAME", funded by the Norwegian Ministry of Environment). Tc-99 discharge data from Sellafield and La Hague were used for model runs. The project "RADNOR" is financially supported by the Research Council of Norway in the Programme "ProFo".

### REFERENCES

- AMAP 1998: Chapter 8: Radioactivity (eds: P. Strand et al.). AMAP Assessment Report: Arctic Pollution Issues. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway, 525-619.
- Brown, J.E., Iosjpe, M., Kolstad, K., Lind, B., Rudjord, A. & Strand, P. 2002: Temporal trends for <sup>99</sup>Tc in Norwegian coastal environments and spatial distribution in the Barents Sea. *J. Environm. Radioact.* 60, 49-60.
- Chen, Q., Aarkrog, A., Nielsen, S., Dahlgaard, H., Lind, B., Kolstad, A. & Yu, Y. 2001: Procedures for determination of 239, 240 Pu, 241 Am, 237 Np, 234, 238 U, 228, 230, 232 Th, 99 Tc and 210 Pb 210 Po in environmental materials. *RISØ Report* Risø-R-1263(EN), 33 pages.
- Dahlgaard, H. 1994: Sources of <sup>137</sup>Cs, <sup>90</sup>Sr and <sup>99</sup>Tc in the East Greenland Current. *J. Environm. Radioact.* 25, 37-55
- Dowdall, M., Gerland, S., Karcher, M., Gwynn, J., Rudjord, A. & Kolstad, A. in press: Optimisation of sampling for the temporal monitoring of technetium-99 in the Arctic marine environment. *J. Environm. Radioact*.
- Dowdall, M., Lind, B., Gerland, S. & Rudjord, A.L. 2003: Geostatistical analysis as applied to two environmental radiometric time series. *Environm. Monitoring & Assessment* 83, 1-16.
- Gerdes, R., Karcher, M., Kauker, F. & Köberle, C. 2001: Predicting the spreading of radioactive substances from the Kursk . *Eos Transactions* 82 (23), 253, 256, 257.
- Gerland, S., Lind, B., Dowdall, M., Karcher, M. & Kolstad, A.K. 2003: <sup>99</sup>Tc in seawater in the West Spitsbergen Current and adjacent areas. *J. Environm. Radioact.* 69 (1-2), 119-127.
- Iosjpe, M., Brown, J. & Strand, P. 2002: Modified approach to modelling radiological consequences from releases into the marine environment. *J. Environm. Radioact.* 60, 91-103.
- Karcher, M.J., Gerdes, R., Kauker, F. & Köberle, C. 2003: Arctic warming evolution and spreading of the 1990's warm event in the Nordic Seas and the Arctic Ocean. *J. Geophys. Res.* 108 (C2), 3034.
- Karcher, M.J., Gerland, S., Harms, I.H., Iosjpe, M., Heldal, H.E., Kershaw, P.J. & Sickel, M. 2004: The dispersion of <sup>99</sup>Tc in the Nordic Seas and the Arctic Ocean: a comparison of model results and observations. *J. Environm. Radioact.* 74, 185-198.
- Kershaw, P. & Baxter, A. 1995: The transfer of reprocessing wastes from north-west Europe to the Arctic. *Deep Sea Res. II* 42 (6), 1413-1448.
- Kershaw, P.J., McCubbin, D. & Leonard, K.S. 1999: Continuing contamination of north Atlantic and Arctic waters by Sellafield radionuclides. *Science of the Total Environ*. 238, 119-132.
- Percival, D. & Walden, A. 2000: Wavelet methods for time series analysis. *Cambridge Series in Statistical and Probabilistic Mathematics, Cambridge University Press, Cambridge, UK.* 594 pages.



**Figure 1:** Measured (symbols and dashed lines) and modelled (solid lines) time series for 5 monitoring sites (see text and legend). Data are shown from 1990 onwards only (modelling data start 1971).



**Figure 2:** Comparison of calculated (NRPA compartment model, vertical axis) and experimental data (measurements, horizontal axis).