The dispersion of ^{99}Tc in the Nordic Seas and the Arctic Ocean in the 1990s according to model results and observations

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INTRODUCTION

Discharges of ^{99}Tc from the Sellafield reprocessing plant increased substantially in 1994, and remained significantly higher than levels throughout the preceeding decade. This resulted from the introduction of new waste treatment procedures (Enhanced Actinide Removal Plant - EARP) which allowed stored medium-active wastes to be processed. EARP was designed to remove actinides and particulates, but allowed ^{99}Tc to be released. This produced a well-defined environmental signal, which could be followed as it spread from the Irish Sea to the North Sea and beyond (Kershaw *et al.*, 1999).
We compare model results from a 3-dimensional coupled ice-ocean model which disperses the ^{99}Tc from the source to the Nordic Seas and the Arctic Ocean, with recent measurements of ^{99}Tc levels in seawater. The observations cover the northern part of the Nordic Seas at different years from the late 1990s. Further, we present a comparison of measurements with an assessment box model, which gives average radionuclide levels on a longer time scale.

MATERIALS AND METHODS

Model descriptions

The dispersion of the recent signal of ^{99}Tc is simulated with the 3-dimensional coupled model NAOSIM (North Atlantic-Arctic Ocean Sea Ice Model) of the Alfred Wegener Institute for Polar and Marine Research. The model has been successfully used previously to investigate the circulation of ice and ocean in the Arctic Ocean and the Nordic Seas (e.g. Karcher *et al.*, 2001; Kauker *et al.*, 2002). The same model has also been used in a previous study to calculate the possible spreading of radionuclides from the sunken submarine 'Kursk' (Gerdes *et al.*, 2001). For the present investigation the model is driven with realistic daily atmospheric forcing data from 1979 to 1999. The atmospheric data are based on data from the European Center for Mediumrange Weather Forecast (ECMWF). The model domain covers the Arctic Ocean, the Nordic Seas and the northern North Atlantic north of 50° N. In January 1990 the release of ^{99}Tc is started in the model run for a period of 10 years, using measured monthly discharge data. The tracer is released into the uppermost model layer of the ocean (thickness 20 m) at the North Channel of the Irish Sea.

The NRPA box model for calculation of concentrations of radionuclides is described in detail by Iosjpe *et al.* (2002). It includes dispersion of radionuclides during time, in order to provide a better and more realistic approach than in traditional box modelling. The model includes the processes of diffusion of radioactivity through pore water, re-suspension, bioturbation effects and burial of activity in sediments. Radioactive decay is included in all compartments. A further advantage of box models is that they can be used for studies involving both large distances (>1000 km) and long time scales, up to the order of 10^3 years.

Measurements

The measurements were performed by the NRPA and CEFAS laboratories. For NRPA 50 or 100 litres of sea water are sampled, prefiltred (1 ìm) in order to remove suspended matter. Then, the ^{99}Tc radionuclides are separated from other nuclides by ion-exchange chromatography using AG1-X4 resin (100-200 ìm mesh size, BIO-RAD Laboratories, Calif., U.S.A.) and subsequent separation techniques such as co-precipitation and solvent extraction. The results for seawater have an accuracy of ±4% (Kolstad & Rudjord 2000). The method is described in detail by Kolstad *et al.* (1999) and Chen *et al.* (2001). For CEFAS discrete near-surface (-5m) water samples (100-150 litre) were collected with shipboard pumps for ^{99}Tc analysis. Anion-exchange was used to separate ^{99}Tc prior to beta-counting, using rhenium as the yieldmonitor (Harvey *et al*., 1992). CTD observations were made at all sampling stations and at intermediate points.

RESULTS AND CONCLUSIONS

The distribution of ^{99}Tc in near-surface waters in 1998 and 1999 is shown in Fig 1. The combined analytical error, due to chemical yield and counting statistics, was generally below 5%. Highest concentrations occurred in the Norwegian Coastal Current. Mixing with North Atlantic Water produceed clear labelling of the West Spitzbergen Current, whilst, lowest concentrations were observed in Polar and Arctic Waters. Also data from the Fram Strait and Western Spitsbergen from 2000 and 2001 (see also Gerland *et al.*, in press a and 2002*)* are by now available and used within the comparison. For the box model comparison, data from the Cumbrian waters, the North Sea and the Skagerrak were used (Brown *et al.* 1999; Leonard *et al.* 1998).

The NAOSIM simulation exhibits the major pathways with the Norwegian Coastal Current and the Norwegian Atlantic Current. Both, model result and measurements show that at the northern end of Fennoscandia, a larger portion of the ^{99}Tc radionuclides is transported eastwards into the Barents Sea, whereas a smaller amount moves north towards Spitsbergen. Another separation occurs in the Fram Strait where a part of the flow recirculates southward joining the East Greenland Current (EGC), and part enters the Arctic Ocean with the continuation of the West Spitsbergen Current (Fig 1). A comparison of the modeled surface concentrations with the available observations in the North Sea and along the Norwegian coast shows good agreement. The levels of surface concentrations in the North Sea northeast of the U.K. in the early phase of the enhanced releases in 1994 are in a range of 0.5-1 Bq/m^3 increasing to about 10 Bq/m^3 in 1996. This compares well with observations. In 1999 at the surface the prevailing current system has carried levels of 0.01-0.1 Bq/m^3 as far as the eastern Eurasian Basin and eastern coast of Greenland down to the latitude of Iceland. By then, the highest levels north of Norway can be found in the Barents Sea, and lower concentrations in the Kara Sea, the Fram Strait and the EGC. Measurements of ^{99}Tc levels west of Spitsbergen and at the islands of Bjørnøya, Hopen and Jan Mayen in 2000/2001 agree well with this

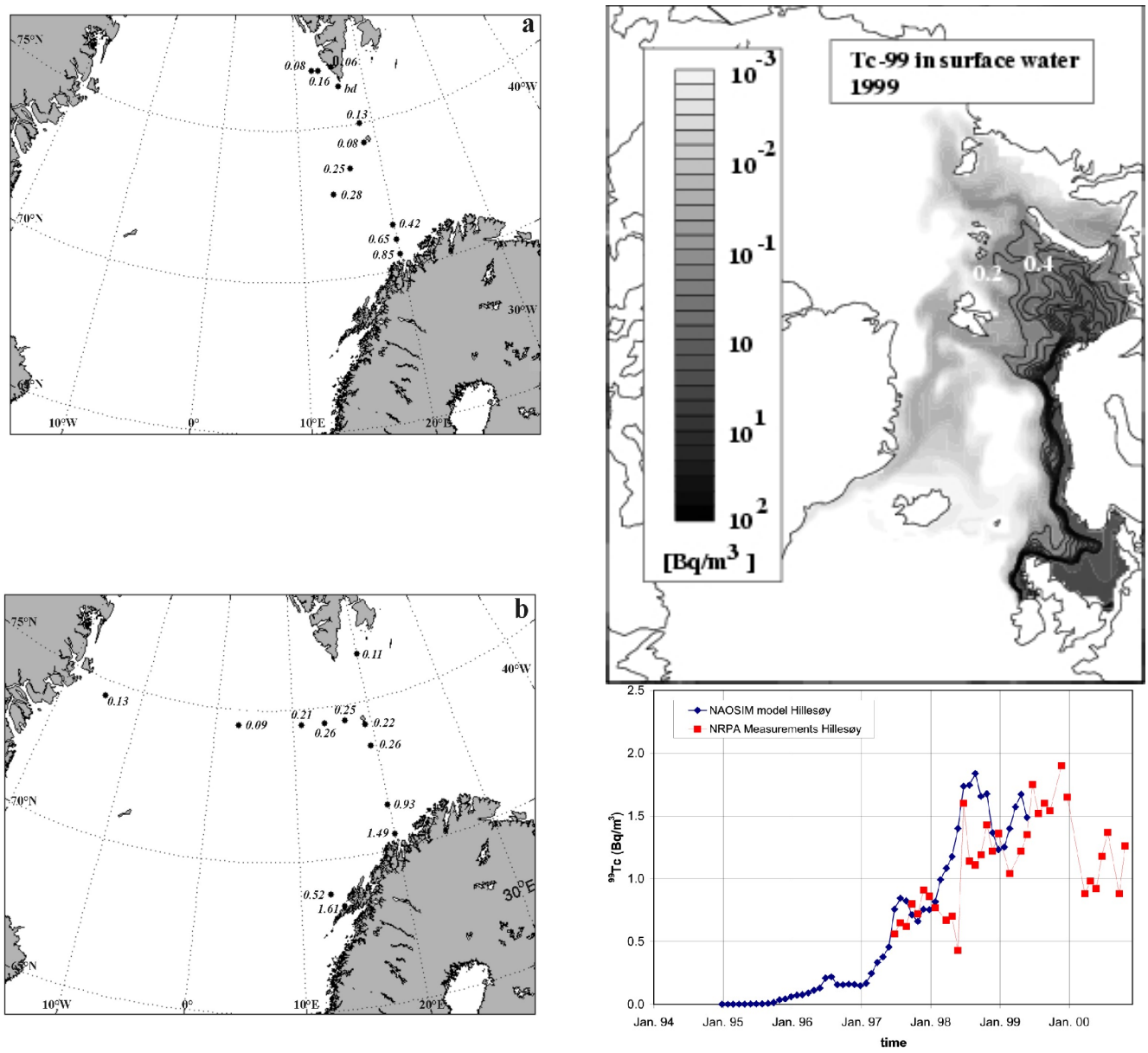

Fig. 1: Distribution of ^{99}Tc (Bq m^{-3}) in surface water in (a) 1998 and (b) 1999, (upper right) NAOSIM model results for 1999 (lines every 0.2 Bq m^{-3}) , and (lower left) timeseries from observations and model results (NAOSIM) near Hillesøy (northern Norwegian coast at 69°N).

picture. The Norwegian Hillesøy time series of ^{99}Tc (Brown *et al.* 2002) agrees well in the magnitude of levels with the model output for the respective area (Fig. 1). Here, at the start of measurements (7-97), the levels reach about 0.6 Bq/m^3 both in the model and in field data. Another strong increase of the ^{99}Tc concentration appears in both model and observation in spring 1998, resulting in levels higher than 1.5 Bq/m^3. The model is approximately three months ahead in relation to the field results. This could be explained with the fact that due to resolution limits the discharge point had to be moved in the model from the real location to

the northern outflow channel of the Irish Sea.
The feasibility of the box model is demonstrated by the correlation of predicted and measured data. Correlation analysis produces an estimate of the correlation coefficient r, of 0.83, the confidence interval being [0.62, 1.03] at the 0.05 significance level. The results and variance analysis show that the box model provides a correct description of the relationship between prediction and measurements for the present data set (not shown).

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