The use of gaseous emissions from a complex industrial site as a means of validating atmospheric dispersion models

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Atmospheric dispersion models are widely used by the nuclear industry from safety planning through to discharge authorisations. This reliance on dispersion models has prompted BNFL to commission Westlakes to assess the accuracy with which models such as NRPB-91 and UK-ADMS can predict air concentrations of radioactive and non-radioactive gases released from the Sellafield site in Cumbria.

A programme of field measurements, which began in 1996, has been undertaken to measure the concentrations of the radioactive gases ^{85}Kr and more recently ^{41}Ar released from the site during routine operations. The resulting databases, which include comprehensive meteorological measurements and time correlated discharge rates now provide extensive validation "toolkits" with which to determine the accuracy of model predictions for a large complex industrial site.

This paper describes the results of the comparison between model predictions and observational data and ranks model performance for a variety of model configurations, meteorological conditions and discharges scenarios.

1 INTRODUCTION

It is increasingly the case that the decision to grant or deny a discharge authorisation or planning application is based on the predictions of an air quality model. However, few ever ask the question, *how accurate are the models?* In turn it could be argued that the modelling community have yet to decide how to measure model reliability, since it can be one or all of the following:

- 100% of predictions are within a certain factor of the true values (e.g. ground level air concentration, deposition rates etc.);
- predictions are most likely to over-predict true values;

- the models are easy to use and their predictions verifiable against hand calculations;
- the input data is accessible and reliable;
- the model performs 'well' during model-model inter-comparisons;
- OR simply…...the model and its configuration are appropriate for the situation.

The nuclear industry use dispersion models for a variety of purposes ranging from routine radiological discharge authorisations and safety planning, through to IPPC assessment of non-radiological emissions. The application of models such as NRPB-R91 (Clarke 1979) and UK-ADMS version 3 (Carruthers *et al.* 1994, CERC 1999) to the BNFL Sellafield site must in some way account for the following complexities:

- multiple discharge points;
- complex array of buildings; and
- close proximity to coastline.

This reliance on dispersion models has prompted BNFL to commission Westlakes to assess the accuracy with which models predict air concentrations of radioactive and non-radioactive gases released from the Sellafield site. A programme of field measurements, which began in 1996, has been undertaken to measure the concentrations of the radioactive gases ^{85}Kr and ^{41}Ar released from the site during routine operations. The resulting databases of field measurements are correlated with comprehensive meteorological observations and discharge data to provide an extensive model validation "toolkit".

2 FIELD MEASUREMENTS

2.1 Krypton-85 (^{85}Kr)

The noble gas ^{85}Kr is a beta/gamma emitting radionuclide (half-life = 10.7 years) produced from the fission of uranium. It is released during the reprocessing of spent fuel at the THORP and MAGNOX plants on the Sellafield site. Approximately 100,000 TBq are released from Sellafield per year, however, the radiological impact of this radionuclide is small and the predicted adult critical group dose is 1.3 μSv a^{-1} (BNFL 1999).

Samples of air were collected in PTFE bags at numerous locations downwind of the emission points. The air samples were returned to the Westlakes laboratory for crude separation of the total krypton fraction (stable krypton + ^{85}Kr) using a cryogenic distillation technique. Fine separation of the krypton fraction and beta counting to determine the ^{85}Kr content were performed by the Radiation and Environmental Physics Group at the University of Gent in Belgium (Hill *et al.* 2001). Table 1 provides a summary of the sampling programme.

Table 1 Summary of the ^{85}Kr field measurements

Site	number of samples	Air concentration ($Bq\ m^{-3}$)		
		Mean	Max	Min
Beckermet	1	3	3	3
Calder Farm	24	186	1527	1
Greystoke	8	7	17	1
Lady Wood	33	68	798	1
Met Compound	30	117	701	2
Mid Tarn Farm	6	52	219	11
Seascale	2	7	10	4
Seascale Hall Farm	37	174	1543	1
Westlakes	14	57	282	1
Short term samples	56	504	5510	1
All	211	218	5510	1

^{85}Kr discharge rates from the THORP and MAGNOX stacks were measured directly and the results made available by BNFL for each sampling period. Meteorological observations were obtained from Sellafield's 48 metre met. mast which is equipped with anemometers, wind vanes and thermistors located at eight heights on the mast. From these observations parameters such as wind speed, wind direction, Pasquill stability category, Monin-Obukhov length and precipitation rates were derived for input into the NRPB-R91 and UK-ADMS dispersion models.

2.2 Argon-41 (^{41}Ar)

The noble gas ^{41}Ar is present in aerial emissions from the Calder Hall reactors on the Sellafield site during routine operation. ^{41}Ar is a short-lived ($t_{1/2}$ = 1.83 hours) beta/gamma emitting radionuclide, produced by neutron activation of ^{40}Ar in the Calder Hall reactor cooling air. In 1999 the annual discharge of ^{41}Ar was 2,600 TBq (BNFL 1999), which corresponded to a predicted adult critical group dose of 42 $\mu Sv\ a^{-1}$, which accounts for up to 65% of the total adult terrestrial critical group dose from discharges. ^{41}Ar external dose rates were measured using an Exploranium GR-320 γ-ray spectrometer located adjacent to the residence of the Calder Hall reactor critical group, which is within 1 km of the reactor stacks. Gamma-ray spectra were collected for the period 14th September 2000 to 18th February 2001, however, monitoring was stopped prematurely due to the outbreak of Foot and Mouth disease, which prevented access to the site. However, during this period over 8,000 gamma-ray spectra were collected over a period of 3,634 hours, resulting in an average measured air kerma rate of 5.5 $nGy\ h^{-1}$ (Lowles *et al.* 2001). The absorbed dose in air was converted to an effective dose of 4.0 $nSv\ h^{-1}$ using a conversion factor ($Sv\ Gy^{-1}$) that relates absorbed dose in air as a function of the initial photon energy (Simmonds *et al.* 1995). Data on the magnitude of emissions of ^{41}Ar from the four Calder Hall reactors were obtained directly from the BNFL reactor physicist. Although not measured directly, the generation of ^{41}Ar from Calder Hall was reliably calculated from the known neutron flux and thus emission rates were estimated as a function of reactor power. The power output of the Calder reactors does not vary significantly throughout the day or night, therefore this method was considered to provide a reliable estimate of the ^{41}Ar discharge rates.

Meteorological observations for the monitoring periods were once again obtained from instrumentation located at BNFL's met. mast.

3 MODEL VALIDATION

3.1 Comparison of ^{85}Kr observations with predictions from NRPB-R91 and UK-ADMS

Several statistical tests were performed to assess how well the models NRPB-R91 and UK-ADMS predicted the observed ^{85}Kr air concentrations. These tests included the R^2 statistic which described the scatter of the modelled doses about a linear regression line. The Normalised Mean Square Error (NMSE) test, which was used to quantify the scatter in modelled values about the true measured data (rather than about a regression line) and therefore provided information on the precision of the results. The Mean Bias (MB) was calculated as the ratio of the mean of the measured dose rates to the mean of the predicted. The mean bias therefore represented the over-prediction or under-prediction of the model, as it compares the mean of all the valid measured data to the mean of all the valid modelled results. Finally to assess the accuracy of the predictions, the fraction of modelled values within factors of 2, 5 and 10 of the measured values were calculated.

In total 10 model configurations were tested. These included the buildings, terrain and coastline modules of UK-ADMS, the use of additional meteorological input parameters (e.g. the standard deviation in wind direction and the wind speed at stack height), as well as effective stack heights, derived from previous wind tunnel studies, rather than physical heights. The table below lists the model configuration that performs best against the statistical tests for a variety of situations, which included a comparison against all the field data, for certain meteorological conditions or for specific receptors (e.g. critical group locations).

Table 2 Summary of the relative model performance

Scenario	Optimum Configuration
All data	**UK-ADMS + Buildings Module + Varying Roughness Domain + σ(wind dir.)**
Critical Group location 1	**UK-ADMS + H_{eff} + Coastal Module**
Critical Group location 2	**NRPB-R91 + H_{eff} + U_{stack} + σ(wind dir.)**
Unstable met. conditions	**UK-ADMS + Buildings Module + Varying Roughness Domain + σ(wind dir.)**
Neutral met. conditions	**UK-ADMS + H_{eff} + Coastal Module**
Stable met. conditions	**NRPB-R91 + Heff + U_{stack} + σ(wind dir.)**

3.2 Comparison of ^{41}Ar observations with predictions from NRPB-R91 and UK-ADMS

Figure 1 below shows graphs of measured dose rate versus predicted, which is based on the air concentrations calculated by NRPB-R91 and UK-ADMS coupled to a single semi-infinite cloud model to generate dose rates (the gamma dose module within UK-ADMS was not used for this study). In this study the Calder Hall Reactor stack heights used in the model configurations are effective stack heights derived from wind tunnel experiments.

In the figure below which uses all the field observations, the straight line on the scatter plots represents a perfect fit between the measured and modelled data. The figure illustrates that in general both models over-predict dose rate, but that UK-ADMS performs better than NRPB-R91. This is reflected in the statistical analysis which shows that UK-ADMS over-predicts the dose by 17% (MB=1.17), whilst NRPB-R91 over-predicts by 60% (MB=1.60). In addition, the F2, F5 and F10 statistics show that UK-ADMS out-performs NRPB-R91 with 57% of the predictions from UK-ADMS being within a factor of 2 of the measurements compared to only 35% from NRPB-R91.

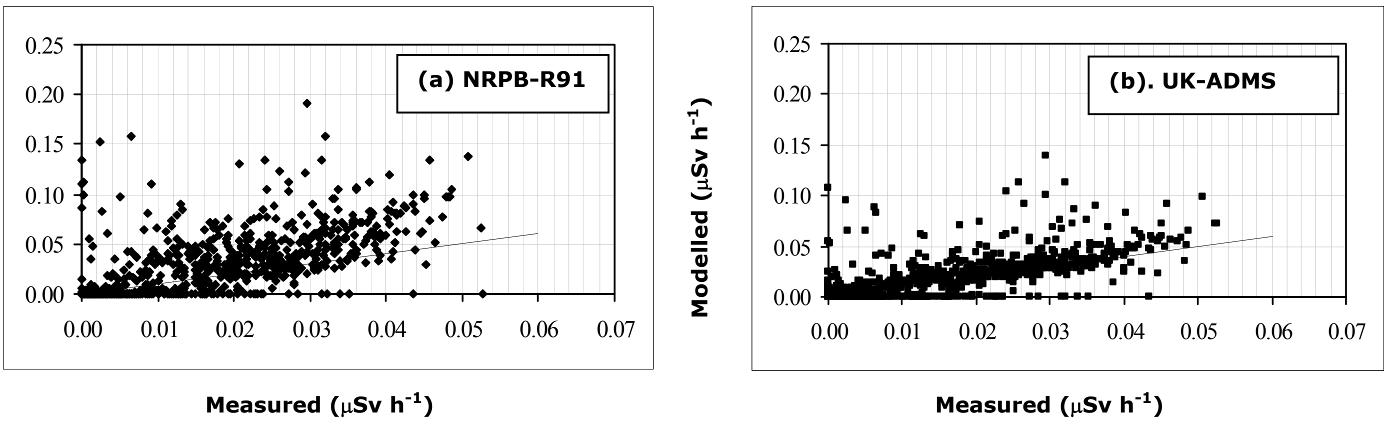

Figure 1 Plots of measured ^{41}Ar dose rates (μSv h^{-1}) versus predicted using EAS and UK-ADMS

The database of measurements and predictions can be interrogated further to investigate those parameters and conditions that influence the calculated dose rates. For example, during neutral stability conditions, when dispersion is dominated by mechanical mixing, both models show a marked improvement in performance with less scatter in the predictions. Data shows that in the case of NRPB-R91, 54% of the predictions are within a factor of 2, whilst 90% are within a factor of 10 and the NMSE is now less than 1.0. For UK-ADMS, 83% of the model predictions are within a factor of 2 of the measurements, whilst almost 100% of the predictions are within a factor of 5 and the NMSE is now 0.27.

4 SUMMARY

The key findings of the study so far are:

- ^{85}Kr and ^{41}Ar field observations have provided an extensive model validation toolkit for the Sellafield site;
- no single model configuration was consistently ranked first - although optimised configurations can be devised;
- for complex sites the effective stack height approach is valid - although requires preliminary wind tunnel experiments;
- UK-ADMS complex effects modules produce results comparable and sometimes better than the effective stack height approach;
- 'Next generation' models are more versatile/reliable if 'applied intelligently'.

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