

ATMOSPHERIC DISPERSION OF RADIOACTIVE RELEASES FROM A NUCLEAR RESEARCH REACTOR: MEASUREMENT AND MODELLING OF PLUME GEOMETRY AND GAMMA RADIATION FIELD

*Bent Lauritzen¹⁾, Helle Karina Aage²⁾, Poul Astrup¹⁾, Kim Bargholz³⁾, Martin Drews¹⁾,
Hans Jørgensen¹⁾, Uffe Korsbech²⁾, Torben Mikkelsen¹⁾, Carlos Rojas-Palma⁴⁾,
Søren Thykier-Nielsen¹⁾ and Raf Van Ammel⁴⁾*

¹⁾Risø National Laboratory, DK-4000 Roskilde, Denmark

²⁾Technical University of Denmark, DK-2800 Kgs. Lyngby, Denmark

³⁾Danish Emergency Management Agency, DK-3460 Birkerød, Denmark

⁴⁾Belgian Nuclear Research Center (SCK•CEN), Boeretang 200, B-2400 Mol, Belgium

INTRODUCTION

In October 2001 a double tracer atmospheric dispersion experiment was carried out at the BR1 research reactor at the Belgium Nuclear Research Center (SCK•CEN) (*Drews et al.*, 2002). The experiment was a collaboration between Nordic Nuclear Safety Research (NKS) and SCK•CEN. A visible aerosol tracer was mixed with the routine emissions of ⁴¹Ar from the air-cooled reactor and released from its 60 m tall reactor stack. Simultaneous measurements were performed of the gamma radiation field from the decay of ⁴¹Ar and of the aerosol plume geometry, in addition to the ⁴¹Ar source term and the main meteorological parameters.

Data from approx. 6 hours of continuous reactor operation were collected during the period October 1-5, 2001. Additional 6 hours of radiation data were obtained without the aerosol tracer measurements. The main wind advection directions during the measurement periods were more or less constant, the wind speeds lower than 6 m s⁻¹ and the estimated atmospheric turbulence predominantly stable to neutral.

In this paper, the experiment is described and main results presented. All measurement data have been analyzed and arranged in a database that can be used to test atmospheric dispersion and dose rate models. It also serves for development of data assimilation methods for atmospheric dispersion of radioactive releases. Aerosol dispersion parameters and the primary photon fluence rates from ⁴¹Ar decay are compared to the results of short-range atmospheric dispersion and dose rate models used for nuclear emergency preparedness. Good overall agreement is found between model calculations and measurements of plume geometry and radiation field.

Experimental setup

During the experiment, the reactor output was kept constant at 700 kW. At this effect atmospheric air is led through the reactor at a rate of 9.4 m³ s⁻¹ giving rise to a constant ⁴¹Ar emission rate of approx. 1.5 x 10¹¹ Bq h⁻¹. The ⁴¹Ar activity concentration inside the stack is recorded continuously using a plastic scintillator mounted with a photo multiplier. Meteorological observations of wind speed and direction, temperature and precipitation were performed by an array of permanent instruments mounted on the weather mast of SCK•CEN. Observations were recorded every minute and subsequently used to estimate the dispersion scaling parameters for 10-min. intervals.

The exact geometry of the radioactive plume was determined from in-situ measurements using a Lidar scanning technique (*Jørgensen and Mikkelsen*, 1993): a white aerosol tracer consisting of a conglomerate of SiO₂ and NH₄Cl was injected into the bottom of the stack with the result that the aerosol plume emitted from the top of the stack would be well mixed with the argon plume. At various locations downwind 2-d plume cross section profiles were determined by scanning the plume with a pulsed laser beam. Aerosol particle positions were determined from the time

delay of the echo and the strength of the echo can be related to the particle concentrations in the plume (Jørgensen *et al.*, 1997). The Lidar scanning was performed from a minibus and measurements were taken both near the stack (~ 100 m) and further away (~ 400 m), but always over the gamma detectors.

The gamma radiation field was monitored using eight NaI(Tl) detectors supplemented by two high-resolution germanium detectors. Four of the NaI(Tl) detectors had thermally insulated 3"x3" crystals and recorded 512-channel energy spectra every 30 seconds while the other four NaI(Tl) detectors consisted of non-insulated 2"x2" crystals connected to single-channel counters yielding integrated count rates every minute. The NaI(Tl)-detectors were deployed along two lines perpendicular to the main wind advection direction at distances up to 1,500 m from the stack; in one case the detectors were grouped two-by-two along a single line for calibration purposes. On each line, the detectors were placed approx. 100 m apart.

Using the spectral radiation measurements the natural background for each detector position was estimated and the background-subtracted full-energy-peak count rate for ⁴¹Ar decay at 1293.6 keV derived. From the net window count rate, n , the fluence rate is obtained as $\mathbf{j} = n/\mathbf{e}$, where \mathbf{e} is the mean detector efficiency (the peak response). The mean detector efficiency was determined from a ⁶⁰Co calibration experiment. For the non-insulated NaI(Tl) detectors, background count rates and detector efficiencies were determined from inter-calibration with the insulated NaI(Tl) detectors.

The germanium detectors were mounted along the plume centerline or next to the NaI(Tl) detectors, providing further calibration measurements. From the germanium measurements it was determined that the emissions from the reactor contained no measurable traces of other radioactive isotopes apart from ⁴¹Ar.

PLUME GEOMETRY

Individual aerosol plume cross-sections were determined from the Lidar measurements at approx. 4 sec. intervals and subsequently binned into larger averaging periods. Figure 1 shows a typical aerosol plume cross section for a 10-min. averaging period. Also shown is the corresponding 10-min average crosswind concentration profile from a Met-RODOS RIMPUFF atmospheric dispersion model calculation (Mikkelsen *et al.*, 1997).

From the Lidar measurements, horizontal and vertical plume dispersion parameters (\mathbf{s}_y , \mathbf{s}_z) can be derived along with the mean position and elevation of the plume. In Figure 2, measured dispersion parameters for Friday October 5 are compared to two different model results:

- a) the SCK•CEN model, in which Pasquill-Gifford stability classes are determined from the wind speed at 69 m and potential temperature gradient over the range 8 m - 114 m, and the diffusion parameters are calculated for each stability class (Bulyneck and Malet, 1972);
- b) the RIMPUFF model in which the puff growth is calculated based on similarity scaling (Thykier-Nielsen *et al.*, 2002; Astrup *et al.*, 2001). Also this model uses the wind speed at 69 m and the temperature differences between 48 m and 78 m, in addition to an assumed surface roughness length of 1 m for the area in question.

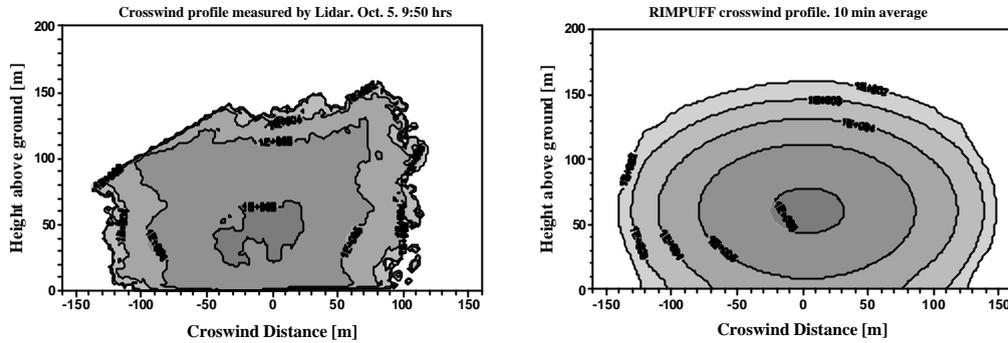


Figure 1. Aerosol plume cross section profile measured approx. 400 m downwind from the 60-m stack by Lidar scanning (left) and the corresponding calculated vertical cross section profile calculated by RIMPUFF (right) for the 10-min averaging period 9:50 to 10:00, Oct. 5, 2001.

Reasonable agreement is found between the measured values and the model results at the downwind range considered, both with respect to the horizontal and vertical dispersion parameters but also to the plume centerline positions. For comparison we also tested the classical Pasquill-Gifford dispersion parameter scheme (Gifford, 1976) but this parameterization yielded significantly smaller values for both σ_y and σ_z . While the site-specific dispersion characteristics were properly picked up by both the site-specific SCK•CEN scheme and the similarity based scheme, the classical diffusion model apparently does not reflect the high roughness and corresponding mixing over the SCK•CEN test site caused by the buildings and tall trees.

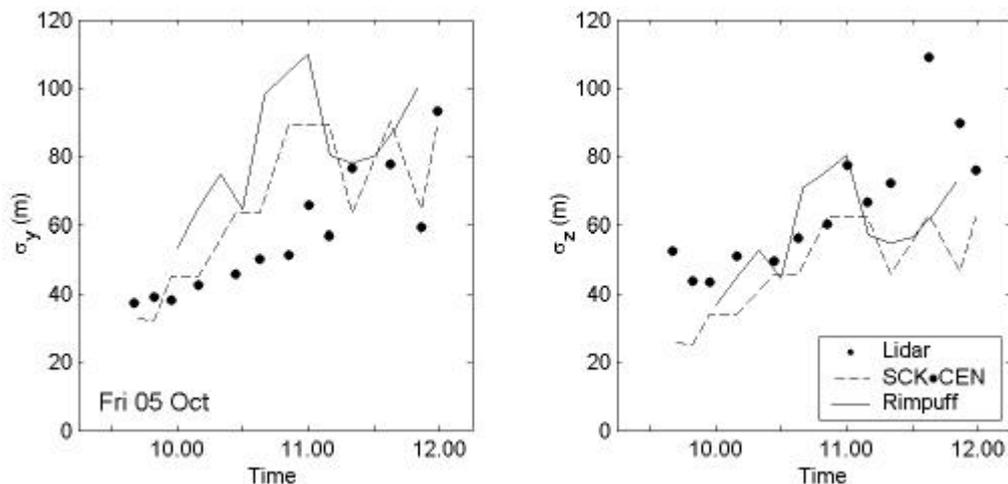


Figure 2. Dispersion parameters obtained from Lidar measurements (dots), from Pasquill-Gifford classification scheme with SCK•CEN parameterization (dashed lines) and from RIMPUFF model (solid lines) using similarity scaling. The increase of the dispersion parameters with time reflects increased turbulence as the wind picked up and the stratification changed from stable over neutral (around 10 o'clock) to unstable in the course of the experiment.

RADIATION FIELD FROM ARGON-41 DECAY

The primary photon fluence rate from the ⁴¹Ar decay is given by

$$\mathbf{j}(\vec{r}_0) = \frac{1}{4\pi} \int d^3\vec{r} \frac{\mathbf{c}(\vec{r}) \cdot \mathbf{y} \cdot e^{-\mu|\vec{r}-\vec{r}_0|}}{(\vec{r}-\vec{r}_0)^2} \quad (1)$$

where λ is the ⁴¹Ar activity concentration in air, y is the gamma-yield per decay ($\approx 99.2\%$) and μ is the linear attenuation coefficient in air ($\approx 0.0069 \text{ m}^{-1}$). The fluence rate is evaluated at receptor points \vec{r}_0 at the locations of the gamma detectors. Because integration is over the entire plume, the fluence rate is less sensitive to variations in the plume dispersion parameters ($\mathbf{s}_y, \mathbf{s}_z$) than direct measurement of the ⁴¹Ar activity concentration λ .

In Figure 3, the primary photon fluence rates recorded by four NaI detectors on Friday, October 5, are compared to the RIMPUFF model. The fluence rate decreases with the distance from the plume centerline (the distance from the average plume centerline to the detectors D, C, A and B being approx. 0 m, 100 m, 200 m and 300 m, respectively). The model calculations are seen to reproduce the overall behavior of the data; the measured fluence rates, however, slightly exceeding the model results. The discrepancy observed shortly after 11.30 can possibly be assigned to the observed break-up of the stable plume due to heat convection, cf. Figure 2.

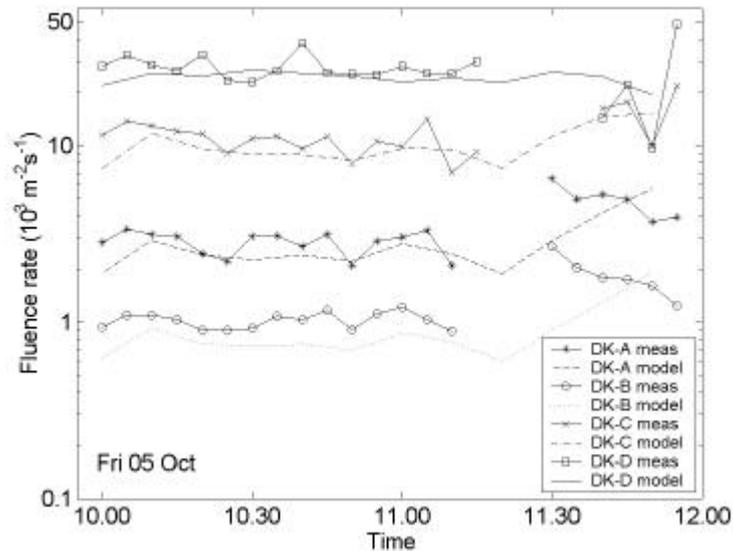


Figure 3. Primary photon fluence rates for detectors DK-A to DK-D compared to Met-RODOS RIMPUFF model calculations.

CONCLUSIONS

Data from an atmospheric dispersion experiment at the BR1 research reactor in Mol, Belgium have been analyzed and arranged in a database suitable for evaluation and development of atmospheric dispersion and dose rate models for nuclear emergency preparedness.

Atmospheric dispersion model calculations of crosswind mean concentration profiles have been compared to direct measurements and reproduce well the data, both with respect to the Lidar measured dispersion parameters and the gross radiation field measurements.

The measured fluence rates, though, are found on average to exceed the model predictions by a factor up to two, and possibly even more at large distances from the plume centerline. This could be due to offset wind directions, or, alternatively, to difficulties in discriminating between the weak fluence rates and background noise levels on the fringes of the plume.

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