Note

MODELING OF THE DISPERSION OF DEPLETED URANIUM AEROSOL

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Fn1 *Abstract*—**Depleted uranium is a low-cost radioactive material that, in addition to other applications, is used by the military in kinetic energy weapons against armored vehicles. During the Gulf and Balkan conflicts concern has been raised about the potential health hazards arising from the toxic and radioactive material released. The aerosol produced during impact and combustion of depleted uranium munitions can potentially contaminate wide areas around the impact sites or can be inhaled by civilians and military personnel. Attempts to estimate the extent and magnitude of the dispersion were until now performed by complex modeling tools employing unclear assumptions and input parameters of high uncertainty. An analytical puff model accommodating diffusion with simultaneous deposition is developed, which can provide a reasonable estimation of the dispersion of the released depleted uranium aerosol. Furthermore, the period of the exposure for a given point downwind from the release can be estimated (as opposed to when using a plume model). The main result is that the depleted uranium mass is deposited very close to the release point. The deposition flux at a couple of kilometers from the release point is more than one order of magnitude lower than the one a few meters near the release point. The effects due to uncertainties in the key input variables are addressed. The most influential parameters are found to be atmospheric stability, height of release, and wind speed, whereas aerosol size distribution is less significant. The output from the analytical model developed was tested against the numerical model RPM-AERO. Results display satisfactory agreement between the two models.**

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INTRODUCTION

THE USE of depleted uranium (DU) in the battlefield provoked a renewed interest in the fate of radioactive aerosol released upon the impact of radioactive material on targets and subsequent fires. Calculating exposures, doses, and health effects on military personnel and the public present in the vicinity of the release is hampered by lack of information regarding input parameters required for quantitative modeling of the exposure. The difficulties arise from the nature of the problem, since studies simulating the occurrence of such a release are only possible by controlled experiments performed by the Armed Forces. Such studies have been performed mainly in the United States, but only a limited number of the reports are publicly available. This is highlighted in the extensive survey of the available literature conducted for the needs of the Royal Society report on the health hazard of depleted uranium munitions (The Royal Society 2001). The Royal Society report focuses on the health AQ: A hazard arising from DU penetrator impact on hard targets and encompasses comprehensively all available knowledge on the issue.

A fundamental characteristic of the problem is the instantaneous nature of the release in the atmosphere in the form of a DU aerosol cloud. Therefore, dispersion of the material cannot be calculated with Gaussian plume models because the latter describe continuous releases. Even when one considers the case of DU burning in fires following the impacts, it cannot be generally described as a continuous release since most of the amount of the aerosol mass is released within only the first 2 h after impact and this only if the fire is externally sustained (Elder and Tinkle 1980). Additionally, because DU aerosol is a material of high density, it is expected to have high deposition velocities resulting from large aerodynamic equivalent diameters. Hence, the simplest physical picture to consider should refer to an instantaneous point release (puff release) with simultaneous deposition. It was not possible to find a working formula for such a dispersion problem in standard texts (e.g., Seinfeld and Pandis 1998; Wicker and Schultz 1982), nor was it possible to find a similar case in the usual compendiums of atmospheric dispersion (e.g., Hanna et al. 1982). Therefore, one has to resort to numerical

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models, which are not readily available. It is, therefore, desirable to identify a working formula that would enable the estimation of such cases. Simple estimations may be sufficient in practice. For the particular application, due to the large uncertainties with respect to the required input parameters, advanced numerical calculations do not necessarily warrant better accuracy. Dispersion of DU aerosol cloud resulting from penetrator impact and fires has been integrated in numerical dispersion calculations to estimate radiation doses downwind of the impact sites using advanced tools like HOTSPOT, AWE-DIFFEL and COSYMA (see, for instance, Liolios 1999; Ferret and von Hippel 1999; Synodinou and Papazoglou 1999; The Royal Society 2001). However, as eloquently discussed in the recent Royal Society report (2001), the reported calculations are characterized by unclear assumptions, whereas ten-fold discrepancies in the final predictions are not uncommon when inter-compared.

The objectives of the present work are two-fold. First, to provide an approximate analytical solution for the case of a puff release with simultaneous deposition. Second, to demonstrate the derived solution by calculating the diffusion and deposition of the released DU cloud as a function of distance and time by employing realistic DU aerosol size distribution and typical weather patterns. Sensitivity analysis of the key input variables that are characterized with the highest uncertainty is performed. The applicability of the proposed approximate analytical solution is assessed by comparing with the predictions of the RPM-AERO code, which is an advanced version of the standard EPA-approved numerical model RPM (Georgopoulos and Seinfeld 1989), which incorporates aerosol processes and heterogeneous chemistry (Lazaridis et al. 2000).

Depleted uranium bulk and aerosol properties

DU is a by-product of the process used to enrich natural uranium ore for use in nuclear reactors and in nuclear weapons. It is distinguished from natural uranium by differing concentrations of certain uranium isotopes. Natural uranium has a ^{235}U content of 0.7%, whereas the content of ^{235}U in DU is depleted to about one third of its original content (0.2–0.3%). The U.S. Nuclear Regulatory Commission (NRC) defines depleted uranium (DU) as uranium in which the content of the ^{235}U isotope is less than 0.2%. The military specifications designate that DU used by the U.S. Department of Defense contains less than 0.3% of ^{235}U. Like natural uranium, DU is a radioactive, heavy metal that emits ionizing alpha, beta, and gamma radiation. Its specific activity is about 12,500 Bq g^{-1} and the half-life is very long (4.5 billion years); therefore, the level of radioactivity does not change over human lifetime.

The aerosol size distribution resulting from DU penetrator impact on hard targets and burning fires has been determined in a number of experiments reproducing to some extent the possible events of aerosol release from DU munitions. Such studies range from the simplified experiment of aerosol production from a DU rod in a tube heated at high temperature (Coleman and Schwendiman 1962) to well contained release studies in enclosed target structures (Chambers et al. 1982). Available literature data indicate that if a lognormal distribution is assumed, mass median aerodynamic diameters (MMADs) for DU aerosol released from impact ranged from 0.8–4.2 μm and geometric standard deviations (GSDs) from 1.8 to 18 (Hansen et al. 1974; Glissmeyer and Mishima 1979; Patrick and Cornette 1978; Chambers et al. 1982; Brown 2000; The Royal Society 2001). The extreme GSD values observed in some of the above studies make the concept of a single mode size distribution inappropriate for such data, which most probably represent many overlapping size modes. Results from studies on DU aerosol released from fires suggest that MMAD ranges from 4–10 μm and GSD from 3.6 to 1.7, respectively, although it is unclear which MMAD corresponds to which GSD (Elber and Tinkle 1980). For the present study aerosol size distributions were selected from the above data. In order to take into account the great uncertainty involved, two enveloping cases were considered. The distribution of 3.7 μm MMAD and 3.5 GSD reported by Brown (2000) is in broad agreement with other results from aerosol impact release studies (Hansen et al. 1974) and aerosol combustion release studies (Elber and Tinkle 1980) and is taken as the reference size distribution. The case of 10 μm MMAD and 1.7 GSD from Elber and Tinkle (1980) is significantly different and is considered as an extreme estimate of the larger aerosol that may be released. DU aerosol mainly consists of two oxides, UO_2 and U_3O_8. Their mass ratio varies in DU aerosol studied in previous reports (Royal Society 2001) across the aerosol size spectrum.

Model development

We consider a point source at height H above the ground, which at time $t = 0$ instantaneously releases an amount of mass corresponding to an activity Q_0. The released puff material consists of aerosol particles that are advected along the horizontal direction x by a wind of

constant velocity u and simultaneously undergoing diffusion in all three directions—x (downwind), y (crosswind), z (vertical)—and deposition along the vertical coordinate z. The upper boundary of the domain is considered to extend to $z = +\infty$; hence, mixing height effects are ignored from the analysis.

Our problem gives rise to the differential equation:

$$\frac{\partial C}{\partial t} + u\frac{\partial C}{\partial x} = K_{xx}\frac{\partial^2 C}{\partial x^2} + K_{yy}\frac{\partial^2 C}{\partial y^2} + K_{zz}\frac{\partial^2 C}{\partial z^2} + v_d\frac{\partial C}{\partial z}, \tag{1}$$

where C is the concentration, $\mathbf{K}$ is the eddy diffusivity vector (with components K_{xx}, K_{yy}, and K_{zz}) and v_d is the deposition velocity. The boundary and initial conditions are as follows:

$$C(x, y, z, 0) = Q_0\delta(x)\delta(y)\delta(H) \tag{2}$$

$$C(x, y, z, t) = 0 \; x, y \rightarrow \pm\infty \text{ and } z \rightarrow +\infty$$

$$K_{zz}\frac{\partial C}{\partial z} = v_d C \; z = 0$$

where δ is the Dirac delta function. The above system of equations does not seem to be amenable to an analytical solution. However, suitable approximations can be made that enable the development of a closed form solution. The most common is the so-called "source depletion" approximation (Slade 1968; Hanna et al. 1982). In this approach the fixed source term in the solution obtained in the absence of deposition is replaced by an effective depleted source term to account for the depletion of the airborne material due to deposition. Accordingly, the time evolution of the effective source term $Q(t)$ can be expressed as

$$\frac{dQ(t)}{dt} = -\int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty} F_s(x, y, t)dxdy \tag{3}$$

with the obvious initial condition

$$Q(t = 0) = Q_0, \tag{4}$$

where t is time and F_s the surface deposition flux. The deposition rate F_s can be expressed in terms of the immediate ground-level air concentration, $C(x,y,z = o,t)$, and the deposition velocity v_d as

$$F_s(x, y, t) = v_d C(x, y, z = 0, t) \tag{5}$$

with the concentration field determined now in the absence of deposition. On the basis of the Gaussian puff formula in an infinite fluid, along with considering the image solution technique to account for the presence of the ground, the solution in the absence of deposition reads

$$C(x, y, z, t) = \frac{Q}{8(\pi t)^{3/2}(K_{xx}K_{yy}K_{zz})^{1/2}} \exp\left[-\frac{(x-ut)^2}{4K_{xx}t}\right]\exp\left[-\frac{y^2}{4K_{yy}t}\right] \cdot\left[\exp\left[-\frac{(z-H)^2}{4K_{zz}t}\right] + \exp\left[-\frac{(z+H)^2}{4K_{zz}t}\right]\right]. \tag{6}$$

Eqns (3)–(6) give rise to a first order differential equation whose solution can be obtained after some tedious but straightforward algebra. The result is

$$Q(t) = Q_0 \exp[-V_d \times I(t)] \tag{7}$$

$$\text{where } I(t) = \left(\frac{4t}{\pi K_{zz}}\right)^{1/2}\exp\left(-\frac{H^2}{4K_{zz}t}\right) - \frac{H}{K_{zz}} \times erfc\left[\frac{H}{(4K_{zz}t)^{1/2}}\right]. \tag{8}$$

The so-derived depleted source term can be now inserted in eqns (5) and (6) to obtain an approximate solution for the deposition flux and air concentration in the presence of deposition.

The numerical evaluation of the above formulae requires the eddy diffusivity coefficients K_{xx}, K_{yy}, and K_{zz} and the deposition velocity v_d to be specified as input parameters. The latter is approximated by the gravitational settling velocity, as determined according to the Stokes law

$$v_d = \frac{D_p^2\rho_p g C_c}{18\mu}, \tag{9}$$

where D_p the particle diameter, ρ_p the particle density, g the acceleration due to gravity, C_c the slip correction (Cunningham) factor, and μ the viscosity of the air. Hence, v_d is dependent on particle size. For a polydisperse aerosol the surface deposition flux F_s is obtained as a weighted sum of the surface deposition fluxes corresponding to each particle size bin of the mass lognormal distribution. It is well known that eqn (9) is a good approximation for large particle sizes (>1 μm), whereas at smaller sizes underestimate deposition. Still, eqn (9) is appropriate for our purposes because in the considered DU aerosol the sub-micron mass fraction is very small (MMAD significantly above 1 μm). The eddy diffusivity coefficients are calculated with the help of the standard deviations across the three directions as follows:

$$\sigma_x^2 = 2K_{xx}t, \; \sigma_y^2 = 2K_{yy}t, \; \sigma_z^2 = 2K_{zz}t. \tag{10}$$

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The above relationships come from the equivalence between the Lagrangian and Eulerian approaches for atmospheric dispersion following a puff release. Contrary to the abundant literature on continuous plume σ's, very few studies exist for the case of an instantaneous puff release. In the present calculations σ's for plume have been used. More specifically, the standard Pasquill-Gifford curves (correlations in the form of power-law expressions) have been employed, as available in standard texts (e.g., Seinfeld and Pandis 1998). In Fig. 1, the correlations employed are compared to some available experimental data on puff diffusion (Hanna et al. 1982). The experiments have been conducted in various atmospheric conditions and circumstances, including artillery bursts. As can be seen, the employed correlations compare fairly well with the experimental data.

RESULTS AND DISCUSSION

The analytical solution derived above is applied for a unitary release of $Q_0 = 1$ Bq. Note that throughout the study it is assumed that the specific activity of the aerosol is uniform, so mass and activity distributions are considered to be identical.

The effect (uncertainty) of key variables is first investigated by performing a parametric sensitivity analysis. The results are presented in Fig. 2a, 2b, and 2c for variations in DU size distribution, release height, and wind velocity, respectively, and for two atmospheric stability conditions. As can be seen in Fig. 2a, the influence of the size distribution is not important within the examined range. Under unstable conditions the particles are deposited closer to the release point. Under stable conditions particle deposition occurs somehow

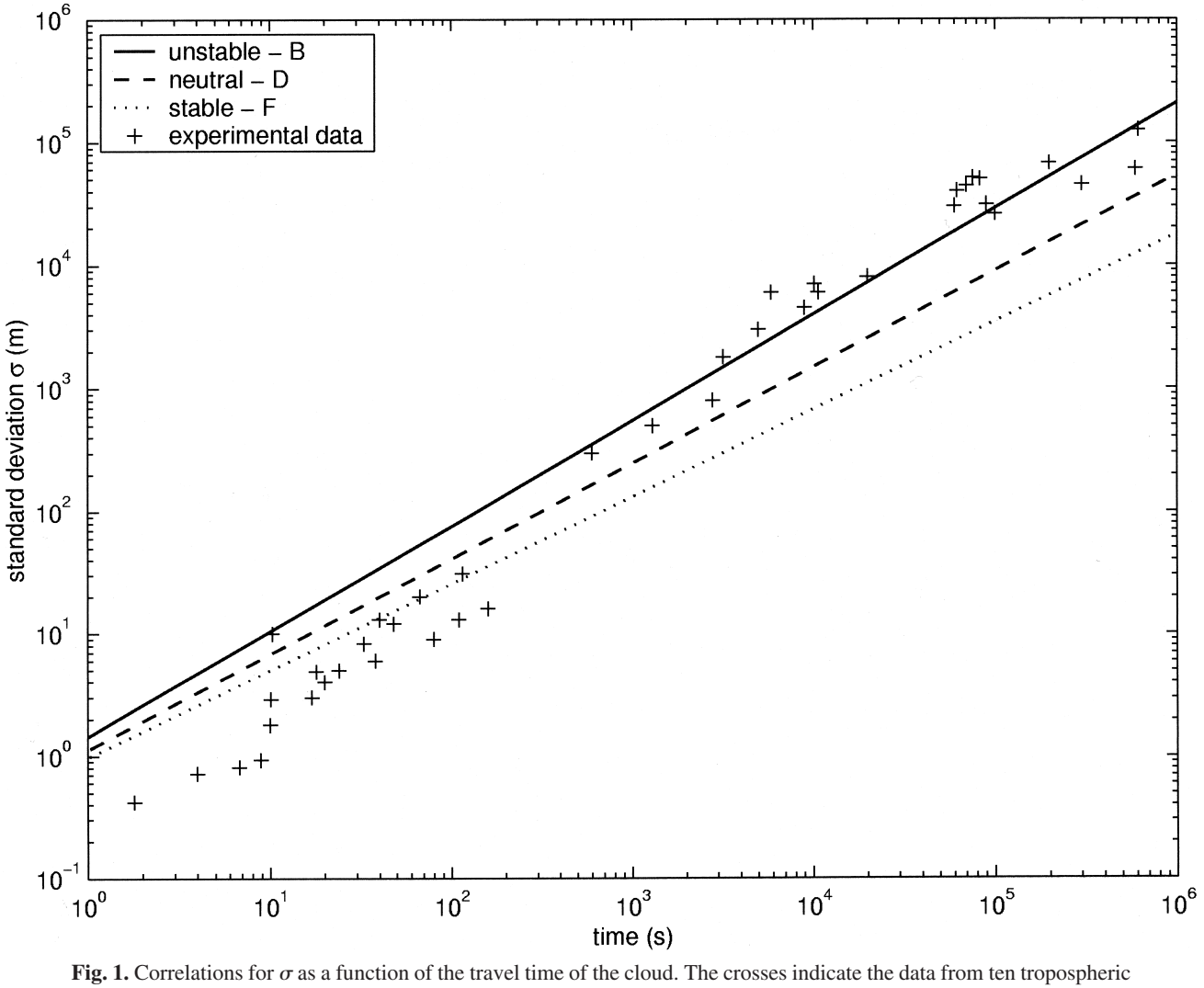

Fig. 1. Correlations for σ as a function of the travel time of the cloud. The crosses indicate the data from ten tropospheric experiments on relative diffusion (Hanna et al. 1982, and references therein).

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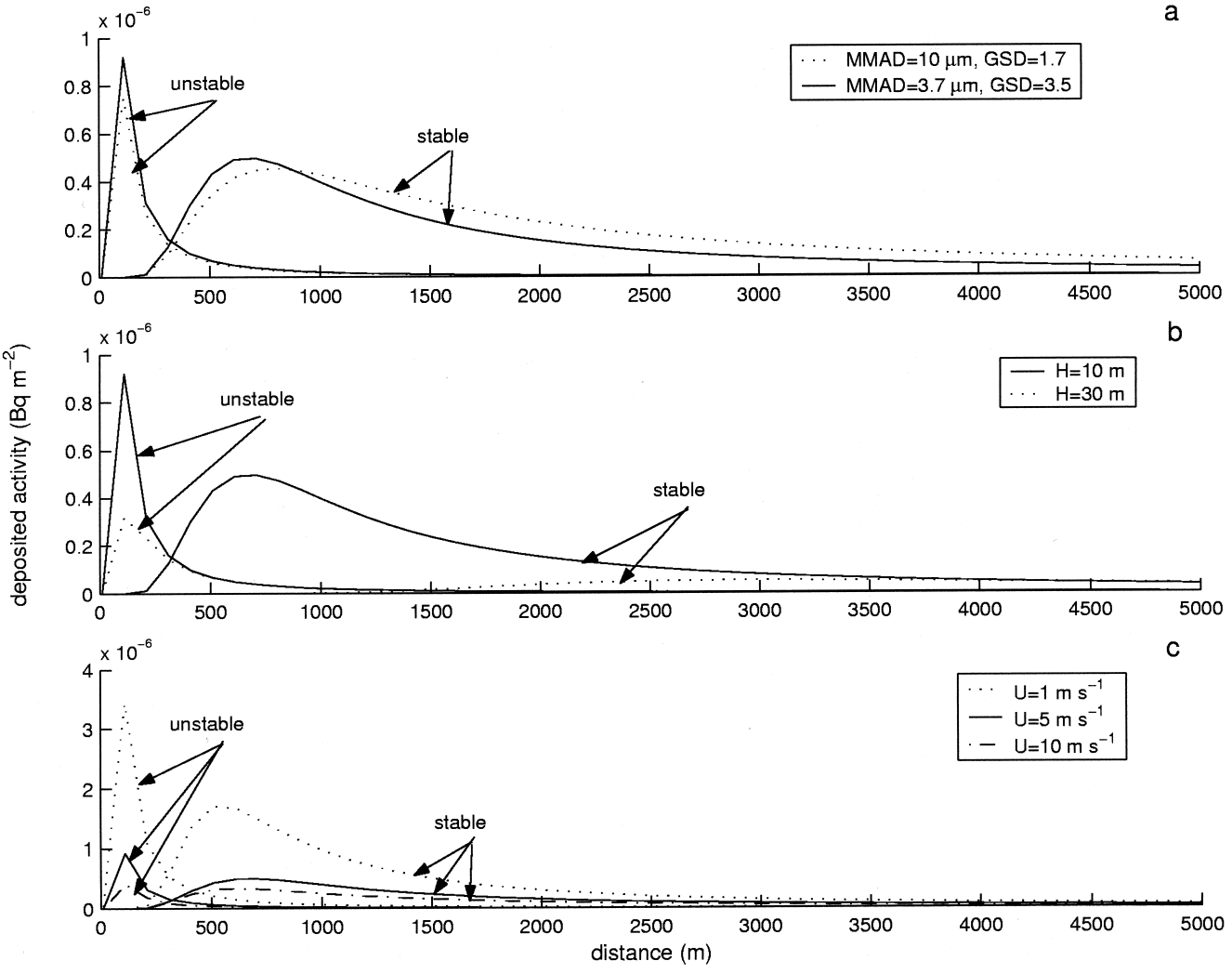

Fig. 2. Total deposited activity on ground as a function of downwind distance from the release point, for different atmospheric stability conditions and different parametric variations (reference parametric values: MMAD = 3.7 μm, GSD = 3.5, u = 5 m s^{-1}, H = 10 m).

further downwind—however, still near the release point. Despite the overall small effect, there is a clear trend to obtain greater deposition for the distribution with MMAD = 3.7 closer to the release point. This must be attributed to the slightly higher mass fraction for the very large particles (>30 μm), which deposit first when GSD = 3.5 compared to the case with GSD = 1.7. Fig. 2b shows the variation of the deposited activity for three different heights of release. Under stable conditions the height of release is a parameter causing a marked effect up to a few kilometers from the release point. Under unstable conditions the considered variations in the release height have negligible effect on deposition after the initial few hundred meters. The third plot (Fig. 2c) shows that, as expected, the ambient wind speed significantly influences the aerosol cloud transport. As can be seen, the lower the wind speed the higher the deposition flux (up to approximately one order of magnitude for the cases examined). In fact, an increasing wind speed promotes advection of particles; hence, deposition flux is less favored. Finally, all plots of Fig. 2 indicate that atmospheric stability has a noticeable influence on deposition, with unstable conditions favoring higher fluxes but more concentrated near the origin; whereas under stable conditions the obtained pattern corresponds to a spread of less contamination but over a larger area.

The time evolution of the air concentration at ground level and 3 levels aloft is shown in Fig. 3 for different distances downwind the release. The characteristic rise and fall in DU air concentration is observed, corresponding to the passage of the cloud from the respective location. Hence, with a puff model one may estimate the period of exposure at any point downwind the release, which is of great importance for radiological

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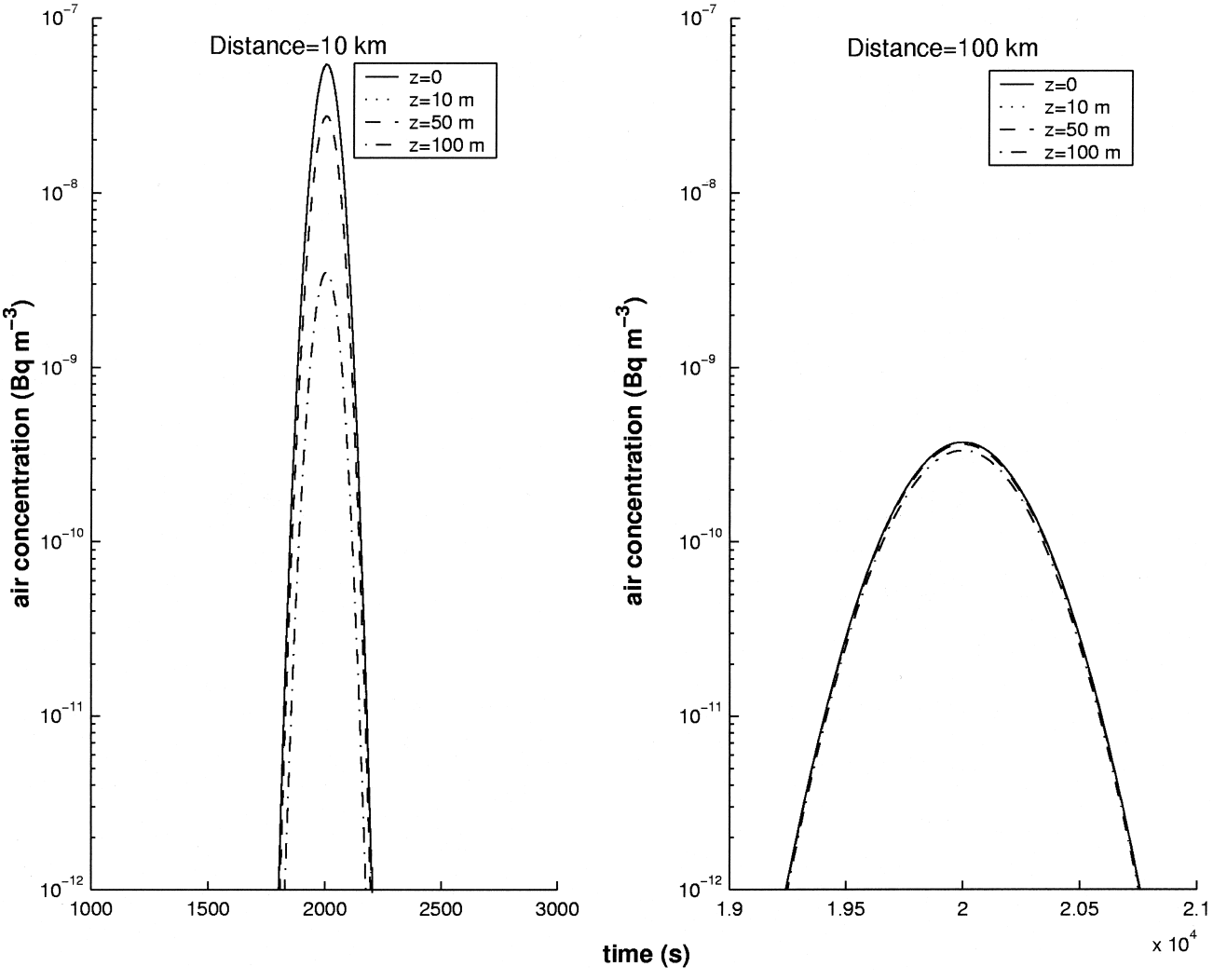

Fig. 3. Air concentration time evolution at different heights and different downwind distances (stable atmosphere, MMAD = 3.7 μm, GSD = 3.5, u = 5 m s^{-1}, H = 10 m).

assessment purposes. Clearly, this feature cannot be replicated with a continuous release (plume) model, and this illustrates the advantage of applying an appropriate puff model solution in the present investigation. The results of Fig. 3 indicate that activity concentration displays vertical variations, which tend to disappear at longer distances downwind. The results reflect the approximations incorporated in the Gaussian model calculations, such as uniform wind speed profile.

In order to assess the performance of the approximate analytical solution developed, the numerical model RPM-AERO was used. The deposited activity calculated by the two models is presented in Fig. 4, as a function of distance. The agreement between the two models can be considered as satisfactory if one takes into account the assumptions incorporated in the analytical model and the discrepancies observed between previously reported results (Royal Society 2001).

CONCLUSION

An analytical solution is derived that accommodates deposition in a puff model. The solution was implemented for the case of depleted uranium particles using realistic size distributions for the released aerosol.

In all the examined cases the activity deposition occurs mainly within a region of a few hundred meters, or a couple of kilometers at most, from the release point. The precise pattern depends significantly on wind speed, height of the DU cloud release, and atmospheric stability and less critically on the DU aerosol size distribution. Previous studies have agreed with the present outcome, reporting that very low values of air concentration and deposition have been calculated at medium range distances. The general conclusion is also validated by calculations performed with the advanced numerical model RPM-AERO. The analytical model developed can

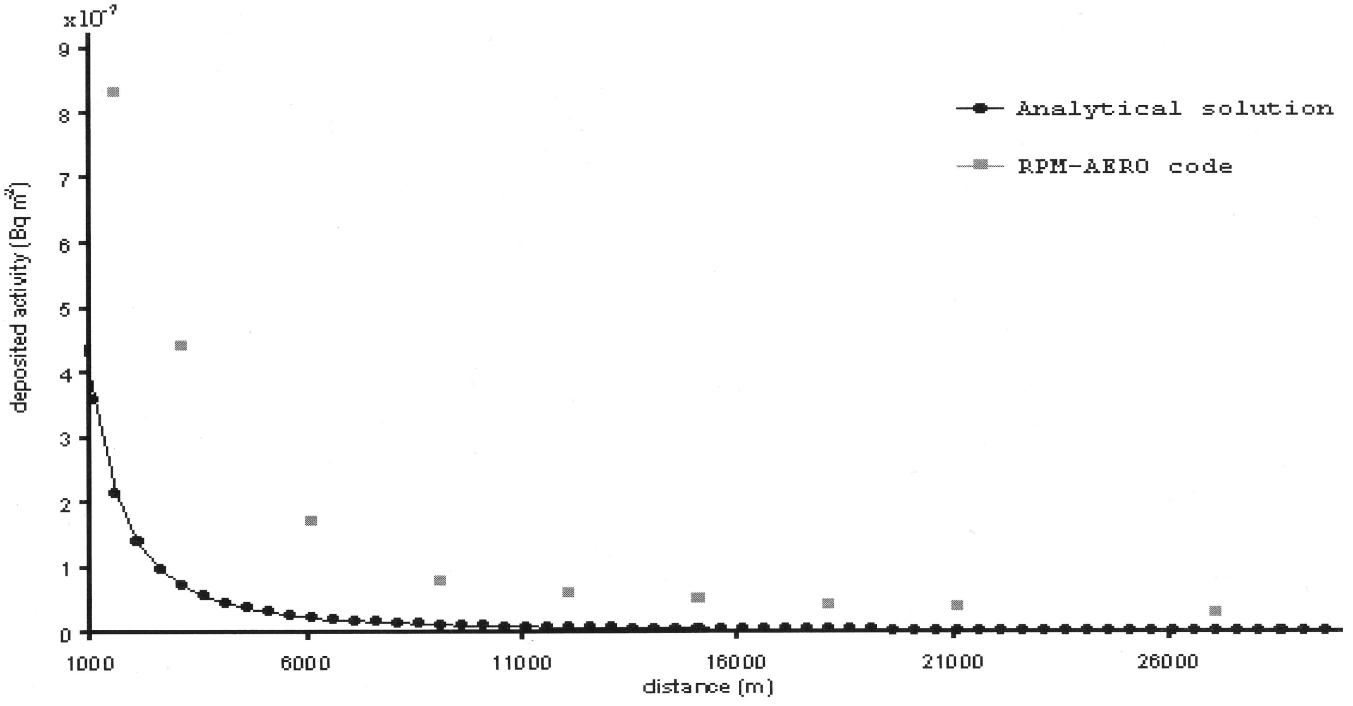

Fig. 4. Total deposited activity on ground, as calculated analytically and with the RPM-AERO code (stable atmosphere, MMAD = 3.7 μm, GSD = 3.5, u = 5 m s^{-1}, H = 10 m).

be applied to exposure estimates in the general case of emergencies resulting from instantaneous releases of radioactive material from other sources.

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