

# Evaluation of accidental radioactive 'atmospheric discharges by nuclear reactors

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## **ABSTRACT**

The primary consideration concerning the safety of a nuclear installation refers to the protection of operators and the surrounding public both during normal operation as well as during an accidental release of radioactive contaminants into the atmosphere. The nuclear electricity generation has proved to be a safe technology, despite the generation of an enormous quantity of radioactive isotopes, both actinides and products of activation, considering the small number of accidental atmospheric dispersion events. In modern reactor installations like that of Angra I in Brazil, the monitoring of radioactivity has not lead to radioactive levels higher than normal accepted environmental levels. This is a consequence of safety features built into the plant in order that, should there be a radioactive release, no members of the public would be subjected to unacceptable levels of risk. Since there have been a very few number of reported reactor accidents, the design of a nuclear plant must resort to theoretical calculations and simulations of hypothetical releases to study the consequences of accidental dispersion of radioactive effluents. In this paper we consider the various stages related with the release of radioactivity from the irradiated nuclear fuel to the calculation of doses, starting from the radioactive inventory calculation followed with the model of radioactive release and the standard treatment for the effluents behavior in the atmosphere suggesting the gamma doses calculation to the public based in the use of the adjoint flux calculations, used as importance function, instead of the direct calculation. This new methodology will be more effective, considering

the inherent large uncertainties in each stage of the radioactive release and dispersion computations. It is also necessary to remember that the system of dose limitation is based on maximum allowed levels of accidental release and maximum pollutants concentration (MCP) on air or on water and in this case the adjoint flux takes the function of the Importance of a particular radioactive release for the dose received by each irradiated individual, or for any other integral response.

## 1. INTRODUCTION

All the nuclear installations, including the nuclear reactors are required to be entirely enclosed by a structure of one or another type to contain radioactivity. The use of multiple, successive barriers to the escape of radioactivity is basic for the design of nuclear power plants. In order to assure that none of these barriers is compromised as the result of abnormal occurrences as equipment failure, human error, or natural phenomena, the national and international regulatory organisms adopted a set of design criteria that establishes the design, fabrication, construction, testing and operation that are important to the safety of the nuclear plant.

All the nuclear reactors emit small amounts of radioactivity, mostly fission products gases, during their operation. They may release considerably more radioactivity during the course of an accident, therefore it is necessary to be able to calculate the doses to the public from such accidental releases, in order to evaluate the environmental impact of a normally operating plant to assure that is within acceptable standards and to investigate the radiological consequences of plant accidents. The computation of doses play also an important role in determining the acceptability of a proposed nuclear plant site.

The basic criteria for the limitation of accidental releases of radioactivity from nuclear installations were originally formulated during the fifties and the permission levels of radioactivity in breathable air and foodstuff following accidental releases to the environment have remained the basis of accident criteria up to the present time<sup>1</sup>. The evolution of the number of nuclear sites and their location progressively closer to urban areas serves to demonstrate

the need for complex physical models to describe various pathways to man.

When a radioactive effluent is released to the atmosphere the plume or cloud is transported downwind as a result of its diffusion in a 3-D domain under the action of fluctuating wind components. As a result the plume is usually expressed in terms of dispersion parameters, for the Gaussian Plume Model (GPM). The GPM gives, in general, very poor predictions for the ground concentrations. The use of GPM with a wide variety of  $\sigma$ 's corrections in dispersion applications, as basis for nuclear reactors regulatory decisions has been under constant discussion. The adoption of the concept of planetary boundary model implied that GPM, as it is used is basically wrong because it does not take into account the turning of the wind speed with height, as observed under all stability conditions.

Due to the potentially severe consequences of the release of radioactive contaminants there have been scientific interests to improve the precision of hazard prediction of such accidents<sup>2</sup>. With the advent of very fast computer facilities it became possible to use prognostic meteorological models coupled with particle trajectory models to estimate atmospheric dispersion in complex terrain. Lagrangian dispersion models are very well suited to simulate the dispersion process<sup>3,4</sup>. With the increasing availability of fast computing systems the use of meteorological models coupled with a particle trajectory model may become a superior choice for studying dispersion in very complex conditions. Radiation doses estimates from planned and unplanned release of radioactive materials coupled with this very expensive computing systems will probably give better results. Despite the very recent 3-D Lagrangian stochastic model for meso-scale atmospheric dispersion applications<sup>3</sup> resulted more accurate than the GPM prediction, for the atmospheric dispersion the results are still strongly dependent on simple models for the plume geometry, soil reflections and do not take into consideration the release of buoyant gases and solid particles.

Before the dose calculations can be carried out, it is necessary to determine how the concentration of the radioactive effluents varies from point to point following its emission into the atmosphere. For these calculations, it is essential to know which are

the potential releases of radioactivity into the environment or source terms. Several studies<sup>5</sup> have identified strong uncertainties associated with the fission products release and transport of pollutants phenomena into the atmosphere. In the past, the focus of the research and analysis of accidental release of radioactivity has been on detailed physical study of the releasing process and transport of individual isotopes. However, in most cases these issues are being addressed in a very restrict context of individual phenomena and gap in scientific knowledge. The purpose of this work is to address the problem of radioactivity and atmospheric transport to individual dose calculation in a broader context to assess any typical radioactive release. This way, it will be not so important the computation of a precise profile of pollutants because the proposal to use the adjoint function has the advantage of using a function that is insensible to uncertainties in the source term as well as insensible to small changes in transport parameters.

## **2. RADIOACTIVE INVENTORY**

The starting point for the environmental assessment of any nuclear installation is the knowledge of the isotopic content of the radioactive inventory. Thus, it is necessary to identify all the main categories of radioactive nuclides arising from different physical processes. In nuclear reactor plants these are:

- a) The inventory of fission products;
- b) The heavy elements (plutonium and higher actinides) produced by successive neutron capture, starting from the uranium isotopes;
- c) Activation products, resulting from the radioactive isotopes of structural and coolant elements formed by neutron capture in the reactor core surroundings.

The computation of the exact radioactive inventory to be obtained under selected conditions for a single reactor installation is very complex and in the past has been performed with tabulations<sup>1</sup>, but in recent years the use of different computer codes<sup>6,7,8,9</sup> to simulate the physics equations to deal with the radioactive inventories becomes very well known and, at least, two of

these codes<sup>6</sup>, are permanently updated for this purpose. The use of accident analysis code<sup>5,10</sup> to specify the class of chemical compounds, associated with the sensitivity analysis will result in the computation of relative importance of individual elements present in environmental release. The significance indicators for the release fractions are pointing, in order of importance, the most significant elements for the content of the source term: noble gases, I, Cs, Te, Mo, Ru, Ba, Sr, Ce and Pu. Overall, despite the phenomenological uncertainties, the predicted source term, into the environment, does not display a high degree for sensitivity due to the airborne capacity of attenuation and retention mechanisms within the reactor containment.

## 2.1. Fission Product Inventory

The burn-up of the uranium fuel, produces many fission products from the  $^{235}\text{U}$  fission, and also from  $^{239}\text{Pu}$ . This last isotope is formed during the reactor operation by neutron capture in  $^{238}\text{U}$ , and towards the end of the fuel irradiation in a typical PWR reactor the fission and therefore the amount of fission products arising from the  $^{235}\text{U}$  is half of that produced by the  $^{239}\text{Pu}$  irradiation. Therefore the fission product yield, must be integrated over the time, reactor core volume and fuel composition in order to get the total radiation inventory.

The primary products of fission are mainly short-lived decaying by beta and gamma emissions. The radioisotopes of radiological significance are in general the fourth and fifth members of decaying chains as in the case of the very well known  $^{90}\text{Sr}$  and  $^{131}\text{I}$ . The radioactivity of the radioisotopes released in the atmosphere is therefore determined by nearly 100 radioactive chains. The combined effects of these chains has to be evaluated in order to estimate the activity released for each isotope. As a consequence the dose of radiation produced by all the radionuclides must also contain the contributions of short-lived isotopes because of its importance to the external gamma dose.

For the dose calculation due to any fission product we need to know:

- The direct yield from fission,  $(\alpha_i)$ ;
- Source from precursors decaying in the same chain;

- Production and depletion by neutron capture;
- Decay constants,  $\lambda_i$ .

The amount of the  $i_{th}$  fission product at a time  $t$  is given by

$$\frac{dN_i}{dt} = -\lambda_i N_i + \sigma_i \phi N_i + \lambda_{i-1} N_{i-1} + \alpha_i F . \quad (1)$$

The fission rate  $F$  necessary to determine the radioactive inventory for a reactor operating at power of  $P$  (MW), can be calculated for a recoverable energy per fission taken as 200 MeV by<sup>17</sup>:

$$F = 3,13 \times 10^{16} P \quad (\text{fissions} / s) . \quad (2)$$

As a consequence the fission product inventory is directly dependent of the reactor power, which value has an inherent uncertainty in the moment of release. The full fission product inventory includes some 600 isotopes, and a complete treatment of decay, for short and long times, from seconds to hundreds of years will be a considerable computation. From various accident reports it is possible to observe the dominant risk of short lived volatile fission products, resulting essential to estimate the full inventory.

## 2.2. Build-up of Heavy Elements

In this context of radioactive inventory computations, a heavy element is that with mass number greater than 230. The activities of transuranic elements are, usually, produced by successive neutron captures in  $^{238}\text{U}$  and heavier elements. The isotopes to be considered as produced this way are normally considered between  $^{230}\text{Th}$  and  $^{250}\text{Cm}$  that are the most likely to be produced by successive neutron captures or radioactive decay from heavy elements in reactor fuels (thorium, uranium and plutonium). The concentration of any of those transuranic elements within the fuel requires the full set of first order differential equations governing the production and decay rates:

$$\begin{aligned} \frac{dN_{ij}}{dt} = & -\lambda_{ij} N_{ij} - \sigma_{ij} \phi N_{ij} + \sigma_{i-1,j}^{n,\gamma} N_{i-1,j} \phi + \sigma_{i+1,j}^{n,2n} N_{i+1,j} \phi \\ & + \lambda_{i+4,j+2}^{\alpha} N_{i-4,j+2} + \lambda_{i,j-1}^{\beta-} N_{i,j-1} , \end{aligned} \quad (3)$$

where the  $\lambda$ s are the constants of all modes of decaying and  $\sigma$ s for the all the neutron capture cross sections involved in this process. The solution of this set of equations due to the strong coupling, must be by numerical methods. Unlikely the fission products equations the parameters in this case are strongly dependent on the reactor type, due to different neutron spectra presented by different reactor concepts. In a typical inventory of heavy isotopes almost all the actinides are alpha emitters and in comparison with the fission products are biologically more effective per unit of activity.

### **2.3. Activation Products**

The interactions of the neutron flux and the atoms within the reactor core and all the surrounding materials, including coolant, moderator and structural materials and its impurities produce many radioactive materials. They are in general produced by nuclear reactions  $(n,\gamma)$ ,  $(n,2n)$ ,  $(n,p)$  and also  $(n,\alpha)$ . These activation products, in general, do not form complex decay chains, like the actinides, decaying by beta and gamma radiation to stable isotopes.

## **3. DISPERSION OF EFFLUENTS**

All nuclear facilities emit small amounts of radioactivity, mostly fission and activation products, during their normal operation and certainly they release considerably more radioactivity during an accident. The environmental impact of a normally operating facility on the public from such releases is related to acceptable levels of environmental radioactivity in comparison with the natural radioactivity. In case of release from an accident the acceptable standards are related to the radiological consequences of reactor accidents. In both cases the computations will play an important role in determining the acceptability for the choice of a plant site. The impact on the population is defined both by the ALARA philosophy and by dose limitation standards, but before dose calculations can be carried out, it is necessary to determine the concentration space profile of the radioactive effluent following its release into the atmosphere.

Having the possibility to establish the radioactive inventory with a model, as discussed in the previous section, we are

able to predict the amount of each isotope leaking into the atmosphere in any given time period. The treatment for the dispersion of effluents in the atmosphere caused by the turbulent motion of the atmosphere must be preceded by the estimation of the effective height of release. The majority of the theories that are proposed to predict the dispersion of windborne materials are based on the Gaussian plume diffusion model. The structure of flow fields is affected by the entrainment amongst surrounding buildings of various size and shape. As a result of model experiments, the Gaussian plume model has been modified<sup>11,12</sup>. The method of allowing for building entrainment that is preferred is due to the virtual source model, but even so is only approximate and is intended as a general guide.

When the plume is emitted from an elevated source, sooner or later the plume will reach the ground. This effect reflects back the Gaussian plume affecting the pollutants concentration and as a consequence the concentration of effluents must be modified by a mirror image source<sup>13</sup>.

Another effect, very complex, that must be used to correct the Gaussian plume model is the Fallout. The fallout occurs under gravity action and is affected also by dry deposition and by rain and snow conditions.

According to the standard diffusion model the effluent spreads out in Gaussian distributions whose dispersion parameters are dependent on the diffusion coefficient, distance from the source and wind velocity in each coordinates axis. Also on the basis of the available experimental data about meteorological conditions defining turbulence types it was possible to define the dispersion parameters<sup>14,15</sup> that apply strictly to short time scales of release, from minutes to 1/2 hour, corrected to include the duration for a release to be explicitly incorporated into the dispersion equation<sup>16</sup>.

The diffusion model for atmospheric dispersion is not an exact description of the phenomenon. As a consequence the use of empirical dispersion parameters gives reasonably accurate values of the effluents concentration for short distances from the source (<10 km). At much larger distances, however, diffusion theory cannot be expected to be valid. The fluctuation in atmospheric conditions tends to disperse an effluent at long distances in an unpredictable way.

## 4. RADIOACTIVE PLUME DOSE CALCULATIONS

In order to evaluate the environmental impact of radioactive effluents from a nuclear facility it is necessary to compute the doses received by the surrounding human population from these effluents as : (1) an external dose from the radiation emitted from the plume; (2) the internal dose from the inhalation of radioactive gases (iodine and noble gases) and radioisotope received from the ingestion of foodstuffs and contaminated liquid effluents; (3) an external dose from radiation emitted by radionuclides deposited on the ground as a consequence of the fallout.

There are several routes by which, the radioactivity can interact with an individual, resulting in direct dose by external body irradiation and also internal dose from inhalation and ingestion.

### 4.1. External Dose

Considering the possibility of having a complete radioactive environment specified in terms of the content of radioactive isotopes in the air and deposited on the ground, following both normal or accidental releases from the nuclear installation, the problem then is to describe the interaction of this radioactive environment and the dose to individuals or population. The origin of the external dose is related with the  $\beta$  and  $\gamma$  irradiation, produced both in the plume environment and from the materials deposited on the ground. Because of the limited range of  $\beta$  radiation in the atmosphere, usually not more than few meters, the dose will depend mainly on the cloud and ground concentrations near the individuals. There is no satisfactory theory developed to describe the behavior the  $\beta$  radiation near the interface air-individual, but various empirical relations are used to approximately compute the dose from  $\beta$  emitters as:

$$\dot{X} \propto \bar{E}_{\beta} \chi_{\beta} \quad (4)$$

where it is possible to observe dependence of the  $\beta$  mean energy and the concentration of  $\beta$  emitters.

The gamma dose is usually computed assuming the photons as emitted from a Gaussian plume. The photon fluence, at any point, is calculated by integrating the  $\gamma$  sources throughout the volume of the plume. For the calculation of external doses, it is usual

to assume a plume infinitely large; this consideration results in conservative dosimetric responses that are larger than they actually are. According to Lamarsh<sup>17</sup> that external exposure rate is given by:

$$\dot{X} = 1,83 \times 10^{-8} \phi_{\gamma} E \left( \frac{\mu}{\rho} \right)_{air} R/s, \quad (5)$$

where  $\phi_{\gamma}$  photon fluence,  $E$  the gamma energy in MeV,  $\mu$  and  $\rho$  are the air absorption coefficient and density respectively. The photon fluence from a point source emitting  $S$  photons/s with energy  $E$ , can be computed by:

$$\phi(r, E)_{\gamma} = \iiint_{E, r, \Omega} \frac{S(E, r, \Omega)}{4\pi r^2} \cdot B(E, \mu, r, \Omega) \cdot e^{-\mu r} dE, dr, d\Omega \quad (6)$$

where,  $B$  is the build up function.

Considering the atmosphere as being infinite, all the photons are absorbed somewhere, therefore, considering a radionuclide,  $\gamma$  emitter, concentration  $\chi_{\gamma}$  (Bq/m<sup>2</sup>) in the cloud, obtained from the diffusion model adopted it is possible to represent the exposition dose<sup>17</sup> as:

$$\dot{X} = 2,62 \times 10^5 \cdot \chi_{\gamma} \cdot \bar{E}_{\gamma} (R/s), \quad (7)$$

where  $E_{\gamma}$  means photon energy. The equivalent dose is also directly proportional to the concentration of each radionuclide in the plume, thus can be written as

$$H = C_{\gamma} \chi_{\gamma} \text{ Gy/s} \quad (8)$$

Noble gases remain airborne at all times, and it is not necessary to compute its influence in other pathways, however, the iodine isotopes, also gases gradually deposited out from the plume onto the soil and vegetation. The rate at which the iodine deposits from the plume is proportional to its concentration and wind velocity:

$$R_d = \chi_{\gamma} v_d, \quad (9)$$

where  $v_d$  is the deposition velocity, therefore the external dose from radioisotopes deposited out from the plume are also dependent from its concentration.

#### 4.2. Internal Dose from Inhalation

It is very well known that the breathing rate  $B$  ( $m^3/s$ ) is a function of the activity we are engaged, as a consequence the total of a radionuclide integrated to a body by inhalation, must be computed during the time of inhalation. The total activity<sup>17</sup> in an organ as a result of a period of inhalation will be:

$$B \int_0^{ti} \chi R(ti - t) e^{-\lambda(ti-t)} dt \quad (10)$$

where  $R$  is a retention function for each organ and radionuclide and  $\lambda$  is the decay constant. The biological dose is, therefore, also dependent on the radionuclide concentration in air, and is because of this the legal limitation MPC in air for each radioactive nuclide, reflecting the maximum limit of dose by inhalation.

### 5. TRANSPORT OF POLLUTANTS BASED ON ADJOINT EQUATIONS

The analysis of the radioactive pollutant effects is usually made through the calculation of radiation doses, examples of which include the gamma and beta doses for external and internal irradiation. As it was emphasized in this work these dose computations rely on the pollutants density function whose profile is determined by direct transport computations, knowing the source term and the physical conditions in which the release of radioactivity has occurred.

Direct transport computations with the diffusion theory are in general very expensive especially when the effect of a radioactive release has to be computed, in a very realistic way, using large number of variables. It is also very difficult to anticipate the effect of a radioactive release, both in normal operational conditions and in case of an accident. This is due to uncertainties in meteorological data and in the profile of the source term.

The algorithm used for sensitivity studies is in general based on conventional perturbation and variational theories. In this work is proposed to use an extension of the perturbation theory methods based on the use of the adjoint of the concentration field,  $\chi^*$ , following a very well known theory used by Lewins<sup>18</sup> in reactor theory and others<sup>19</sup> treating large class of responses, including temperature<sup>20</sup> fields and pollutants dispersion effects<sup>21</sup>.

Considering the problem of estimating a functional "R" of the radioactive pollutant concentration profile  $\chi(x,y,z,t)$ , when the latter satisfies the well known linear transport equation, or any diffusion approximation concerning a dispersion model for the pollutants atmospheric dispersion.

$$L\chi = S \quad (11)$$

where  $L$  is the transport operator defined by according the diffusion model used for the simulation. Assuming that it is possible to carry out the solution of equation (11), for some meteorological condition, within adequate boundary conditions it is possible the computation of various responses of interest through the computation of the inner product<sup>[1]</sup>:

$$R = \langle \Sigma, \chi \rangle, \quad (12)$$

where  $\Sigma$  is the response function of interest, defined in the phase space to the transport equation. According to the response type under consideration the response function may assume a variety of expressions, capable to represent the internal or external doses and any other integral response. For the moment we limit our attention to functionals linear with the pollutants concentration density  $\chi$ . Let  $R = R(X)$  being a function of several input variables that represent a defined dosimetric problem. Starting from some set of variables, it is possible to compute the result of interest using, equation (12). And it is also possible to compute the response for the first order perturbation due to small changes in input variables by:

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[1] The bracket notation indicating the inner product of a response function and the density function, which in this case corresponds to an integral over the independent variables for which the function product is performed. For finite differences computations, using finite dimensional vectors, the same notation indicates a scalar product of two vectors.

$$R(X) = \langle \Sigma, \chi_p \rangle = \langle \Sigma, \chi_r \rangle + \langle \Sigma, \delta\chi_r \rangle, \quad (13)$$

where  $\chi_p = \chi_r + \delta\chi_r$ .

If the reference system is to be used for the calculation, any changes in the input variables can be computed with an error of first order. A more accurate evaluation for the response can be made if a variational principle is constructed for that response, considering that must be stationary to changes in the input variables.

In order to establish a variational principle for the functional of interest we construct the Lagrangian function adding the functional of interest a multiple of the side condition. Thus the Lagrangian function constructed in this way for the reference system will have the next form:

$$La = R_r + \langle \chi_r^+ (L\chi - S) \rangle. \quad (14)$$

The weighting function  $\chi^+$  plays the role of an undetermined Lagrange Multiplier, measuring the rate of change in the response  $R$  consequent upon changes in the transport equation. The initial consideration of constructing a Lagrangian Function for the response function  $R$ , as a stationary function will result in,

$$\delta La = \langle \Sigma, \delta\chi \rangle + \langle \delta\chi, L^+ \chi^+ \rangle \rightarrow 0. \quad (15)$$

Therefore the undetermined Lagrange multiplier can now be defined by:

$$L^+ \chi^+ = -\Sigma, \quad (16)$$

where the adjoint function  $\chi^+$  plays the role of a Lagrange multiplier. In this context the adjoint function can be considered a second independent distribution that relates the effect of pollutants distribution at space point on observable of interest. The solution of  $\chi^+$  for the distribution will be obtained from the adjoint transport equation, in any of the diffusion models adopted:

$$L^+ \chi^+ = S^+ \quad (17)$$

where  $S^+$  is the adjoint source for any response of interest. We follow, here, the interpretation made by Lewins<sup>18</sup>, as the Importance of each particle in contributing to the detector response. Therefore in the same way the adjoint source is really a particle detector of some kind and can be remembered physically as a particle sink. This

interpretation, resulting for the adjoint as a dual function of the pollutants concentration means that,

$$\begin{aligned} R &= \langle \Sigma, \chi \rangle = \langle S^+, \chi \rangle \\ &= \langle S, \chi^+ \rangle \end{aligned} \quad (18)$$

Considering that the Lagrangian function was constructed as a variational principle for the response  $R$ , the error in evaluating a response functional is of the second order:

$$\delta La \rightarrow \delta R = \langle \delta \chi^+, L \delta \chi \rangle. \quad (19)$$

Therefore the fundamental Lagrangian function gives an estimate of the response  $R$  accurate to second-order in  $\delta \chi$  and  $\delta \chi^+$ .

This approach based on adjoint equations allows us to estimate the extent of the potential danger of atmospheric pollution in a region away from all the sources, with prescribed scenarios of meteorological regime.

As an example, to apply this methodology, let us examine the problem of global transport of pollutants, with the unique aim of discussing the construction of the adjoint operator for the diffusion equation .

### **Example: Global transport of pollutants**

The model adopted for global transport of pollutants is the diffusion equation with a relatively more realistic model in which the medium moves with the wind velocity. the model considers a spherical system of coordinates  $(\theta, \psi, z)$ , where  $\theta$  is a longitude,  $\psi$  is a supplement to latitude and  $z$  is a height counted from the underlying surface. The diffusion equation for the density of pollutants is then:

$$\begin{aligned} &\frac{\partial \chi}{\partial t} + \frac{u}{a \operatorname{sen} \psi} \frac{\partial}{\partial \theta} + \frac{v}{a} \frac{\partial \chi}{\partial \psi} + (w - w_g) \frac{\partial \chi}{\partial z} \\ &- \frac{\partial}{\partial z} v \frac{\partial \chi}{\partial z} - \frac{1}{a^2 \operatorname{sen}^2 \psi} \frac{\partial}{\partial \theta} \mu \frac{\partial \chi}{\partial \theta} - \frac{1}{a^2 \operatorname{sen} \psi} \frac{\partial}{\partial \psi} u \operatorname{sen} \psi \frac{\partial \chi}{\partial \psi} = S \end{aligned} \quad (20)$$

The pollutant concentration  $\chi = \chi(\theta, \psi, z, t)$ , is computed with the wind effect  $\vec{u} = (u, v, w - w_g)^T$ . The turbulence diffusion coefficients in horizontal and vertical directions are  $\mu$  and  $\nu$  respectively. The function  $S = S(\theta, \psi, z, t)$  represents the power and location of pollutant sources, and "a" is the average earth radius.

The approach based on the importance equations allows writing the adjoint equation for the model presented in equation (20) as follows:

$$-\frac{\partial \chi^*}{\partial t} - \frac{u}{a \sin \psi} \frac{\partial \chi^*}{\partial \theta} - \frac{v}{a} \frac{\partial \chi^*}{\partial \psi} - (w - w_g) \frac{\partial \chi^*}{\partial z} - \left( \frac{\partial}{\partial z} \nu \frac{\partial \chi^*}{\partial z} + \frac{1}{a^2 \sin^2 \psi} \frac{\partial}{\partial \theta} \mu \frac{\partial \chi^*}{\partial \theta} + \frac{1}{a^2 \sin \psi} \frac{\partial}{\partial \psi} u \sin \psi \frac{\partial \chi^*}{\partial \psi} \right) = S^+ \quad (21)$$

where  $S^+$  is a function determining the functional represented in the equation (6). Let us use this methodology to determine the amount of radioactive pollutants in a certain sub-domain  $\{T, \Omega\}$ ;

$$R = \int_0^T dt \int_{\Omega} S \chi^* d\Omega = \int_0^T dt \int_{\Omega} S^+ \chi d\Omega \quad (22)$$

As a consequence the amount of radioactive pollutants in a certain sub-domain can be computed using the solution of an adjoint problem  $\chi^*$ . As it was said before, this function plays the role of a weight function affecting the contribution of every radioactive pollutant source  $S$  to the atmosphere pollution over the selected sub-domain. This represents a useful way of making possible to map more or less dangerous zones.

## 6. CONCLUSION

The theoretical models used to study atmospheric dispersion in very complex situations like those of radioactive effluents in normal or in accidental dispersion due to a reactor accidents are very limited in order to compute with a desirable accuracy the biological effects on personnel and members of the public. Comparing the possibilities of the GPM models with more advanced fully 3-D Lagrangian particle trajectory models it was possible to understand the difficulties for both models to represent with a desired accuracy a realistic simulation of atmospheric dispersion under variable meteorological conditions. First is the conceptual difficulty related with the exact definition of the source term under accidental release of radiation and second is the difficulty to model the atmospheric dispersion process. The governing meteorological parameters for atmospheric transport and diffusion vary significantly both in space and time. Hence, the analytical solutions used for atmospheric dispersions, such as Gaussian plume or puff model, may not yield realistic results when used with such meteorological data. With the advent of fast computing facilities, it is possible to use meteorological models coupled with particle trajectory codes, capable to model the dispersion in inhomogeneous and complex terrain conditions. Even so the dose rate estimation using such very complex computing system will be affected by the uncertainties in the source term and in the meteorological data.

In this work we introduce the concept of the adjoint function for the concentration of radioactive elements in a plume as an importance function for the biological effect or dose. This methodology, originally used to study the neutron transport in reactor physics, will prove to be very efficient, when used to study the sensitivity of the radiological effects due to uncertainties in the source term and in the meteorological data. The efficiency of this model is guaranteed by the inherent characteristics of the importance function, as a second independent distribution that relates the effect of particles distribution on the observable of interest. This additional information for estimating the importance function or the source importance to a detector response arises since we know the particles origin that caused the detector response. This means that we can estimate the dosimetric response of a detector at any point  $r$  to a

source  $r'$  for the system considered. This is equivalent to estimate the Greens's function  $G(r \rightarrow r')$  of the system.

It may be noted that if a problem consists of examining the effects of different sources, or plume pollutants distribution, for a fixed detector function, the adjoint source, then it is more efficient to solve the adjoint equation and determine the response by  $\langle S, \chi^+ \rangle$ , even for a very simple model. After the numerical testing with this model it will be very interesting to explore adjoint construction model with the 3-D Lagrangian stochastic models.

## REFERENCES

1. Clarke R.H., MacDonald H.F., Radioactive releases from nuclear Installations: evaluation of accidental atmospheric discharges, *Progress in Nuclear Energy*, 2,2, (1978).
2. Ducros G., Malgouryres P.P., Kissane M., Boulaud D., Durin M., Fission product release under severe accidental conditions: general presentation of the program and synthesis of VERCORS 1-6 results., *Nuclear Engineering and Design* 208, 191-203, (2001).
3. Raza S.S., Avila R., Cervantes J., A 3-D Lagrangian stochastic model for the meso-scale atmospheric dispersion applications, *Nuclear Engineering and Design*, 208, 1-28, (2001).
4. Oza R.B., Panchal N.S., Nambi K.S.V., Krishnamoorthy T.M., Coupling of mesoscale meteorological model with particle trajectory model to study the atmospheric dispersion under sea breeze conditions, *Environmental Modeling & Software*, 16, 63-71, (2001).
5. Ang M.L., et alii, A risk-base evaluation of the impact of key uncertainties on the prediction of severe accident source terms-STU, *Nuclear Engineering and Design*, 209, 183-192, (2001).
6. Bell M.J., "ORIGEN, The ORNL Isotope generation and depletion code "Report ORNL-4628, (1973)
7. England T.R., CINDER, A One point depletion and fission product code, Report WAPD-TM-335, (1962)
8. Clarke R.H., FISP, A Comprehensive computer program for generating fission product inventory, *Health Physics*, 23,565-572, (1972).
9. Harte G.A., HYACINTH A heavy isotope point burn-up and decay code, CEGB Report RD/B/N3564, (1976).
10. Alpert D.J., Chanin D.I., Ritchie L.T., Relative importance of individual elements to reactor accident consequences assuming equal release fractions, Report NUREG/CR-4467, (1988).
11. Gilford F.A. Turbulent Diffusion typing schemes- a review, *Nucl. Safety* 17, 68-86,(1976).
12. Pasquill F. Atmospheric diffusion, Halstead Press, (1974).
13. Csanady G.T., Turbulent Diffusion in the Environment, D. Reidel Publishing Co. Holland, (1980).
14. Pasquill F., The estimation of the dispersion of windborne material, *Met. Mag.*, 90, 33-49, (1961).

15. Smith F.B. A scheme for estimating the vertical dispersion of a plume from a source near the ground level, Report NATO-CCHS 1, (1973).
16. Moore D.J., CEGB Report Surrey England(1976).
17. Lamarsh J.R., Introduction to Nuclear Engineering, 2<sup>nd</sup>. Ed. Addison Wesley, USA, (1983).
18. Lewins J., Importance, Pergamon Press, NY, (1965).
19. Graça C.O. Reaction Rate Studies in a Fusion Reactor Blanket, PhD, Thesis, Cambridge University, England, (1986).
20. Gandini A., Generalized Perturbation Theory Methods. A Heuristic Approach, in Advances in Nuclear Science and Technology, 19, Plenum Pub. Co.,(1987)
21. Marchuk G.I., Agoshkov V.I., Shutyaev V.P., Adjoint Equations and Perturbation Algorithms in Nonlinear Problems, CRC Press Inc., Florida USA,(1996).

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