Quantification of inhaled soot mass using Pathfinder coupled results

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ABSTRACT

To be able to handle the fire risks present at CERN, the HSE unit needs to assess the risks relating to fire involving radioactive materials. Smoke consists of a multi-phase flow, containing combustion gases and solid particles in suspension, what is commonly called soot.
The possible effect of radioactive smoke is quantified using a novel methodology, based on Pursers fractional effective dose (FED) model for toxicity [1]. This new methodology is compatible which is compatible with the output generated by a coupled FDS-Pathfinder simulation. The methodology has been specifically developed to be able to import Pathfinder output files and based on visibility and CO concentration, calculate the mass of inhaled radioactive soot particles and gases. The difference between the phases has been made to consider effects as soot deposition, which would not occur for radioactive gases. This methodology has been verified in several case studies.
This approach allows the evaluation of the incurred Fractional Effective Dose, which is a built-in function of Pathfinder, as well as the related inhaled mass of soot and radioactive gases, which in turn give us the amount of incurred radioactivity.
A case study will be presented showcasing the integrated FDS-Pathfinder approach with the developed methodology added on.

GLOSSARY

Y_s	The soot yield of a fuel (-)
A_i	Total activity of i (Bq)
m_i	Mass of i (kg)
a_i	Specific activity (Bq/kg)
ρ	The local density in the cell (kg/m^3)
K_m	The mass-specific extinction coefficient (8700 m^2/kg), fuel dependent
S	Visibility (m)
C	Non-dimensional constant characteristic of the type of object being viewed through the smoke (8 for light emitting signs, 3 for reflective signs) (-)
χ_i	The mass fraction of i (-)
f_i	The volume fraction of i (-)
W_i	Molar mass of i (kg/mol)
RMV	Volume of air inhaled (l/min)
RMV_{mass}	Mass of air inhaled (kg/s)

INTRODUCTION

CERN, the European Organisation for Nuclear Research, is the world's leading laboratory for particle physics which employs ca. 3700 staff, graduates and fellows full time. Over the course of 2023, CERN trained about 800 students and hosted about 1000 associates. It also provided infrastructure and services to about 12400 users.

CERN provides a unique network of accelerators that collide particle beams head on or direct them onto fixed targets. The flagship machine is the LHC, large hadron collider, which provides colliding beams of protons and other particles at the highest energies ever achieved. The products of these collisions are recorded by the ALICE, ATLAS, CMS, LHCb, LHCf, MoEDAL and TOTEM experiments, and, since 2022, also by the newcomers FASER and SND@LHC. The year 2023 was marked by the second year of Run 3 of the LHC, at an energy of 13.6 TeV, and by the first heavy-ion run in five years, at an energy of 5.36 TeV per nucleon–nucleon collision [2]. As CERN operates a network of accelerators, these collisions also occur in experiments in other facilities such as PS or SPS and their respective experiments (ISOLDE, n-TOF, North Area, etc.).

These collisions and their products emit stray radiation which activates materials in the vicinity of the beam lines. Correct alignment, collimators, dumps and shielding limit the stray radiation, but in many cases, equipment is needed in the vicinity of the beam lines. Most of the equipment which needs to be located close to a beam line are magnets and cables to power these magnets as well as send control signals. Other potentially combustible equipment that can be activated can be targets, beam dumps, vacuum oil pumps, etc.

Given that CERN is constantly increasing the energies of these collisions to discover new particles and new physics, it is possible that an unmitigated increase in collision energy increases the irradiation of the equipment, and therefore formation of unwanted radionuclides, in the vicinity of the beam line. There are several ways in which a fire can lead to releases of radionuclides:

- The combustion of fuel forms soot particles which contain the formed radionuclides.
- Due to the combustion of the fuel, some radionuclides are released as gas or volatile elements or compounds.
- Radionuclides which were formed inside metallic materials could be released through out-diffusion and taken up into a soot particle or released as a gas. [3]

Fire hazards at CERN are therefore connected to a radiological hazard for occupants, intervention personnel, bystanders and the environment. To quantify the risk level involved with these radiological hazards, CERN has developed a methodology which is based on ISO 16732-1, called the FIRIA methodology, which stands for Fire Induced Radiological Integrated Assessment. This has been started as a project and is currently in its second phase, the large-scale implementation to several case studies.

THE FIRIA METHODOLOGY

Methodology development

Given the diverse conditions found at CERN, the FIRIA methodology is continuously being updated [4].

One such major update was in the quantification of the risk of a given facility. Whereas fire frequencies were attempted to be calculated in the first phase, this was shown to be very difficult due to the uncertainty and quality of the input data. Moreover, the formulation of the risk acceptance criteria for the spread of radionuclides are formulated in a way that lean towards a consequence analysis instead of a risk analysis with inherent frequencies.

For occupants who are occupationally exposed workers, a limit of 6 mSv was fixed (category A workers). These limits are in line with the EU and Swiss regulations. However, disregarding the fixed limits, optimization in the sense of the radioprotection ALARA principle is required [5]. It was proven that the inhalation effective dose dominates the total effective dose for persons evacuating from the facility in case of fire.

Only on a secondary level, a full risk analysis is used to determine the cost-benefit of certain mitigation measures – but dedicated studies for proposed mitigation systems are deemed more suitable for this purpose than a risk assessment of a complete given facility.

Mapping knowledge gaps

Even though the existing FIRIA methodology has a lot of tools at its disposal for its successful application, a hiatus was found in the quantification of the occupants' radiation effective dose due to inhalation. A literature study did not find any similar methodologies to quantify the effective dose due to inhalation, using CFD coupled agent-based software. Validated tools have been developed for the calculation of effective doses to bystanders and the public in the environment but so far no methodology has been developed to assess the risk for occupants, who are present in a room with an accidental fire.
A novel approach was chosen which is based on Pursers fractional effective dose model, which is implemented in Pathfinder. By using soot and CO as tracers for the different kinds of radionuclides (solid, liquid, gaseous and volatile), an inhalation effective dose could be calculated for each occupant.

Current built-in capabilities of Pathfinder

At present, the level of integration between FDS and Pathfinder can only trace the following time-dependent properties per agent:

- Temperature
- Volumetric O_2 fraction
- Volumetric CO fraction
- Volumetric CO_2 fraction
- Visibility

These five properties are coupled as FDS PLOT3D output data, which is made available to the Pathfinder simulation as input. It is currently not possible to couple more than these five properties. These properties can then be used to define local slowing down due to visibility [6] and can be used to assess the fractional effective dose (added effects of CO, CO_2 and lack of O_2) [1]. Temperature can be used to define whether some places in the computational domain are impassible due to excessive temperatures. Pathfinder then outputs the data for each specified occupant in the output folder.

THEORETICAL MODEL

As described before, the release of radionuclides from the bound form in the fuel can happen in three forms:

- In the form of solids, liquids or volatile compounds, which are assumed to incorporate or attach to soot particles.
- In the form of radioactive gases and volatile compounds, which do not bind to soot particles but rather behave as gases.
- Through out-diffusion. These radionuclides then either bind to soot or escape with gases as mentioned above.

Soot as carrier for radioactive elements

One of the two pathways is therefore via soot. The FDS modelling used in the FIRIA methodology uses a deposition submodel, so not all the soot leaves the domain or is suspended in the air [7]. Based on the inhalation of soot mass we can calculate the total inhaled activity of soot-bound radionuclides knowing he inhaled soot masses and specific activities. This way we can calculate the inhaled mass of radioactive elements which bind to soot particles.
The inhaled mass of soot can however not be used for the assessment of the amount of inhaled radioactive gases, because soot deposits whereas radioactive gases would not.

As a demonstrative example, consider Sb_2O_3 and I_2. Unstable antimony and iodine nuclides can be formed in fuels which are exposed to a radioactive environment.
It is assumed that all antimony trioxide produced is bound to the soot produced by the fuel, and is suspended with that soot as trace particles. The carrier species, soot, can however deposit on walls and other objects, as the simulations are ran with the deposition submodel activated. Hence, the concentration of suspended soot is lower than without the deposition model activated.
Iodine gas is a gas and as such would not bind to soot particles, hence soot deposition would have no effect on the concentration of gaseous radionuclides.

CO as a tracer for radioactive gases

CO production (as well as soot production) is modelled in the FIRIA methodology through complex stoichiometry, which in essence can be calculated back to a CO yield. In this paper, CO is used as a tracer gas for radioactive gases and volatile substances as it is a gas and, moreover, not present in air in the initial conditions of a simulation. Its molecular weight is close to N_2 and O_2 and therefore it mixes well with air.

Specific activity of soot

To calculate the inhalation effective dose in Sievert due to the inhalation of smoke, it is important to model how the radioactivity in the fuel is transferred to soot. The mass of the produced soot is calculated as:

$$m_s = Y_s m_{fuel} \tag{1}$$

With m_s being the soot mass, m_{fuel} being the mass of burnt fuel, and Y_s the soot yield of the fuel.
We also know that the total activity in the fuel and in the soot is respectively:

$$A_{fuel} = a_{fuel} m_{fuel} \tag{2}$$

$$A_s = a_s m_s \tag{3}$$

With a_i being the specific activity of i.
If we then assume that all the activity in the fuel is transferred to the soot, which is a conservative assumption:

$$A_{fuel} = A_s \tag{4}$$

$$a_{fuel} m_{fuel} = a_s Y_s m_{fuel} \tag{5}$$

$$a_s = \frac{a_{fuel}}{Y_s} \tag{6}$$

This gives us the specific activity of the soot, and using the inhaled mass of soot we can calculate the inhaled activity in Bq. The inhaled activity can be converted to the inhalation effective dose by using dose coefficients [Sv/Bq] precalculated for many radionuclides by the ICRP [8].

Using visibility to calculate soot mass fraction

Since occupants do not stand still during a fire, but rather evacuate, it is not representative to just integrate the soot mass fraction in one point to obtain a total mass of inhaled soot. The methodology should be similar to the one used in the coupling between FDS and Pathfinder.

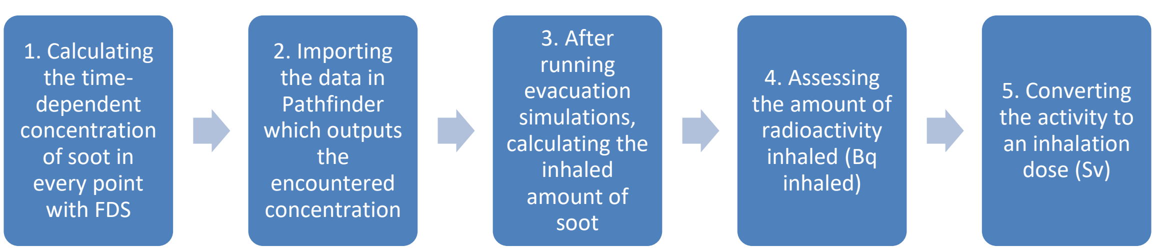

Figure 1. Overall methodology to calculate the inhalation effective dose for occupants.

For our purposes, the amount of inhaled soot is needed. This can be calculated using the soot concentration (mass fraction) . In Pathfinder, there is currently no way of explicitly tracking soot concentration per agent in a 3D simulation.
It is however possible to output the time-dependent visibility encountered by every agent.
Using reverse engineering, it is possible to obtain the encountered soot mass fraction in every point from the encountered visibility.
As stated before, we can use the property visibility S, an output of FDS, as an input in Pathfinder. We know the visibility S at every time step. We also know that:

$$S = \frac{C}{K_m \rho \chi_s} \tag{7}$$

Or:

$$\chi_s = \frac{C}{\rho K_m S} \tag{8}$$

Calculating the inhaled soot mass and activity

In Pathfinder and in Pursers model [1] a volume of 25 l/min is suggested which corresponds to a 70 kg human engaged in light activity over periods of up to one hour (who is walking to an emergency exit).
The simple equation $RMV_{mass} = \rho\ RMV/(1000 \times 60)$, with $\rho = 1.2$ kg/m³, gives us the mass of air inhaled per second: 0.5 g/s.
We also know that there are several gases present in smoke which affect breathing. Of these, CO_2 is the one which absolutely needs considering, as CO_2 greatly increases the RMV, which will increase the of soot uptake and therefore the radioactivity uptake. It is therefore necessary to calculate a multiplication factor (V_{CO_2}) to allow for the effect of the increased RMV caused by carbon dioxide on the rate of soot uptake [1]. The expression for this is:

$$V_{CO_2} = \exp\left(\frac{[CO_2]}{5}\right) \tag{9}$$

In which $[CO_2]$ is the volumetric concentration of CO_2 in %.
The mass of inhaled air per second is then:

$$V_{CO_2} RMV_{mass} \tag{10}$$

Multiplied by the soot mass fraction χ_s, the mass of inhaled air becomes the mass of inhaled soot per second:

$$\chi_s V_{CO_2} RMV_{mass} \tag{11}$$

Or:

$$\dot{m}_{inhaled-soot} = V_{CO_2} RMV_{mass} \frac{C}{\rho K_m S} \tag{12}$$

This can be integrated over timesteps:

$$m_{inhaled-soot} = \sum V_{CO_2} RMV_{mass} \frac{C}{\rho K_m S} \Delta t \tag{13}$$

The activity inhaled with soot is therefore:

$$A_{inhaled-soot} = a_s m_{inhaled-soot} \tag{14}$$

$$A_{inhaled-soot} = a_s \sum V_{CO_2} RMV_{mass} \frac{C}{\rho K_m S} \Delta t \tag{15}$$

$$A_{inhaled-soot} = \frac{a_{fuel}}{Y_s} \sum V_{CO_2} RMV_{mass} \frac{C}{\rho K_m S} \Delta t \tag{16}$$

This assumes that none of the inhaled soot is exhaled, which is again a conservative assumption.
All the variables on the right-hand side of equation (16) are given either as inputs (constants) or as integrated properties between FDS and Pathfinder. The density ρ can be calculated using the ideal gas law:

$$\rho = \frac{\rho_0 T_0}{T} \tag{17}$$

The temperature encountered in each point is also one of the integrated properties between FDS and Pathfinder.

Calculating the inhaled amount of CO and the inhaled activity of radioactive gases and volatile substances

We assume that radioactive gases and volatile substances would behave as CO. As the produced gases have different molecular weights, they eventually would separate due to gravity. However, since these radioactive gases are only present in relatively small amounts, and due to the time scales involved, it can be assumed that turbulence and diffusion will have a larger effect on the gas mixture in the smoke, and therefore CO can be used as a tracer gas.

Contrary to the soot mass fraction, Pathfinder can output the CO volume fraction per time step for each of the agents in the domain. If we assume the molar mass of smoke to be the same as air:

$$f_{co} = \frac{\rho_{smoke}}{\rho_{co}} \chi_{co} \tag{18}$$

$$f_{co} = \frac{W_{smoke}}{W_{co}} \chi_{co} \tag{19}$$

$$\frac{W_{co}}{W_{air}} f_{co} = \chi_{co} \tag{20}$$

Formula (20) allows us to convert the CO volume fraction to the CO mass fraction.

Assuming the ratio between the inhaled activity $A_{inhaled}$ [Bq] and the inhaled CO mass $m_{inhaled\ co}$ [kg] is equal to the gaseous activity liberated from the mass of the burnt fuel $a_{fuel}m_{fuel}$ [Bq] and the mass of the liberated CO, $Y_{\mathrm{CO}}m_{fuel}$ [kg]:

$$\frac{A_{inhaled}}{m_{inhaled\ co}} = \frac{a_{fuel}m_{fuel}}{Y_{\mathrm{CO}}m_{fuel}}$$

and

$$A_{inhaled} = \frac{1}{Y_{\mathrm{CO}}} a_{fuel} m_{inhaled\ co}$$

This finally leads to an analogous formula as the formula for inhaled soot:

$$A_{inhaled} = \frac{a_{fuel}}{Y_{co}} \sum V_{CO_2} RMV_{mass} \left(\frac{W_{co}}{W_{air}}\right) f_{co}\, \Delta t \tag{21}$$

Converting inhaled activity in Bq to an effective dose in Sv

The final model step is to convert the inhaled activity in Bq into the inhalation effective dose in Sv. For each radionuclide, a dose coefficient in Sv/Bq is available [8]. The effective doses due to the different radionuclides are then summed to a total inhalation effective dose.

APPLICATION TO A CASE STUDY

To this date, this methodology has been applied to 3 case studies:

- The FIRIA ISOLDE SZ case study, in which an experimental hall is connected to a separator area for beams, which contains activated and contaminated combustible materials.
- The FIRIA NA TCC2/TDC2 case study, an underground hall where targets and therefore activated fuels, mostly magnet resin and cables, are present. Access of personnel is granted only after several hours of cool down and area air flush following each beam stop.
- The FIRIA PS Ring case study, an accelerator ring where activated fuels are present (mostly magnet resin and cables). Access is not allowed except when the beam is off and access is granted after a given cool-down time.

FIRIA NA TCC2/TDC2 case study

Building Description

The TCC2/TDC2 area is located in the North Area (Figure 2) of the Super Proton Synchrotron (SPS). The building BA80 (building 889) allows people to access to the TDC2/TCC2 (Tunnel 886) area. It is located on the Prévessin site (Figure 3)

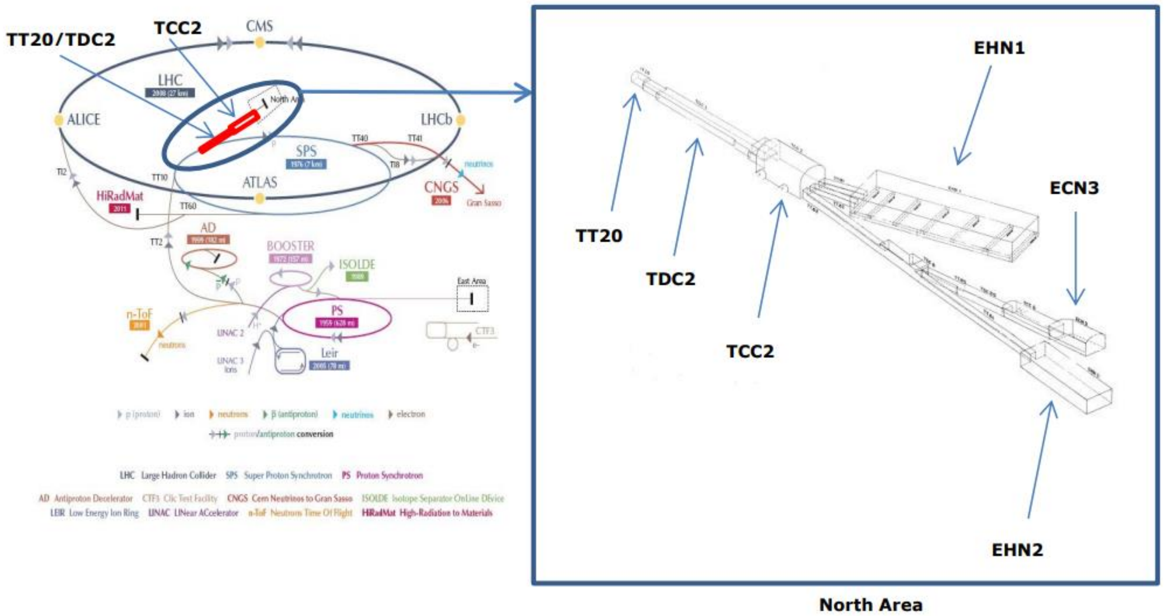

Figure 2. General overview and location of TDC2 and TCC2.

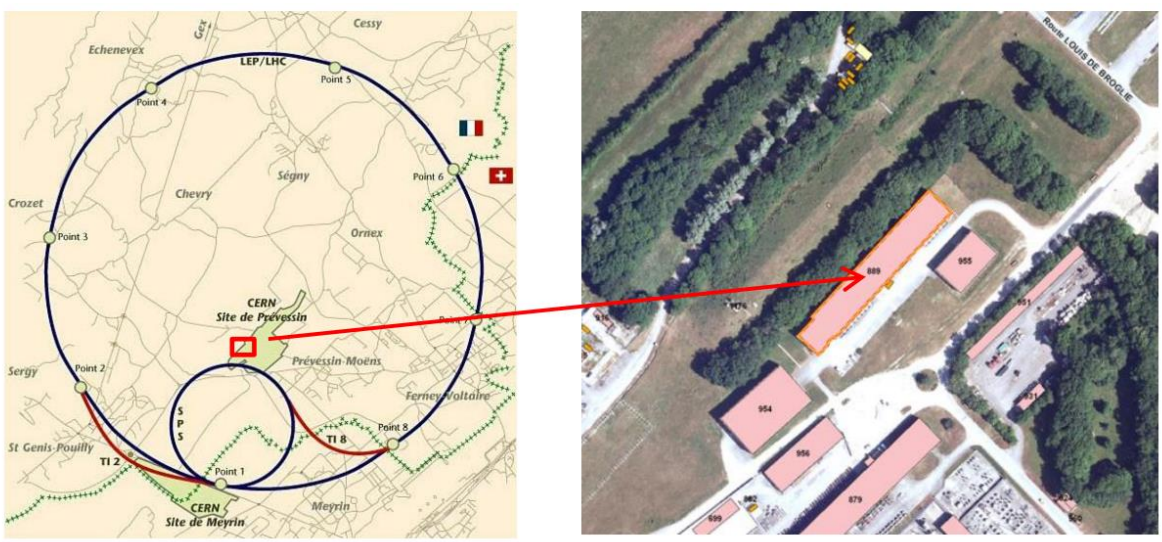

Figure 3. Location of the Prévessin site and BA80, the auxiliary building to access TDC2/TCC2.

The general infrastructure of the TDC2/TCC2 area includes the BA80 building which allows entering the tunnel through one elevator or through a staircase located in the PA80 which leads to the TA801 corridor. This corridor leads directly to the TCC2 area (See Figure 4). The GT802 tunnel is used only as an emergency exit with stairs located in the PGT801 shaft to reach the surface.

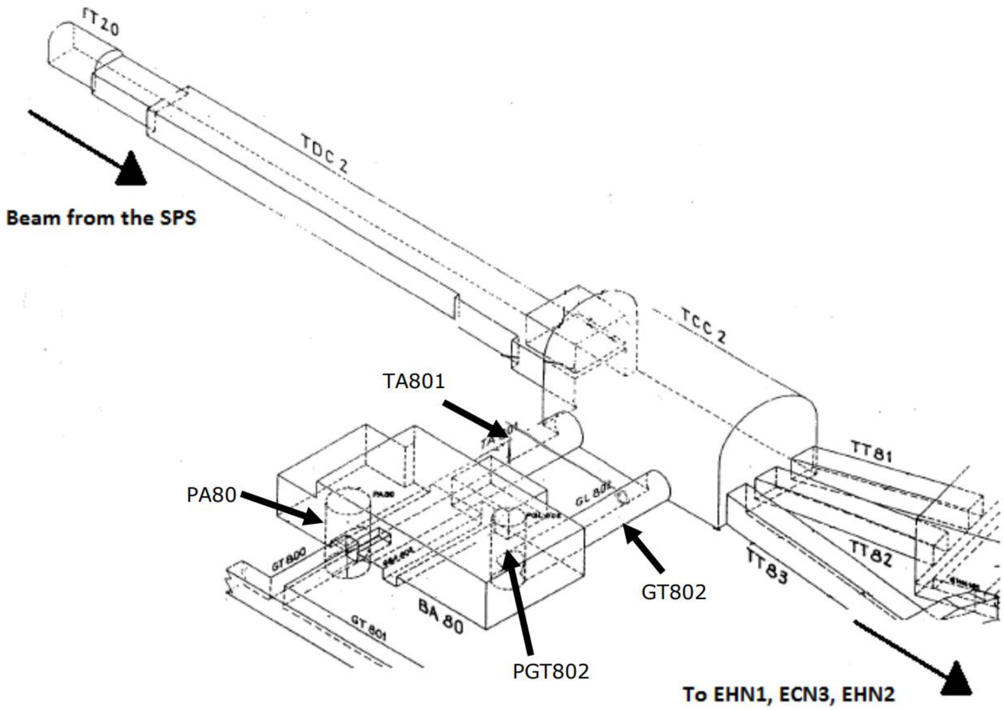

Figure 4. Layout of the TDC2 and TCC2.

The TDC2 tunnel is 228 m long and between 6 m and 10 m wide, with a height of about 4 m. At the junction with TCC2 the height is doubled because of the technical space on the upper floor.
TCC2 is 130 m long, 16 m wide and 10 m high.
In the junction area TDC2/TCC2, a second floor is present. This ventilation room (Vent room) contains all the ventilation equipment.

Effective dose due to inhalation

For evacuating occupants, the inhalation dose is proportional to the inhaled soot mass and inhaled CO mass , which are dependent on the pre-movement times and the total evacuation times. Figure 5 [9] shows nomenclatures for different parts of an evacuation analysis.

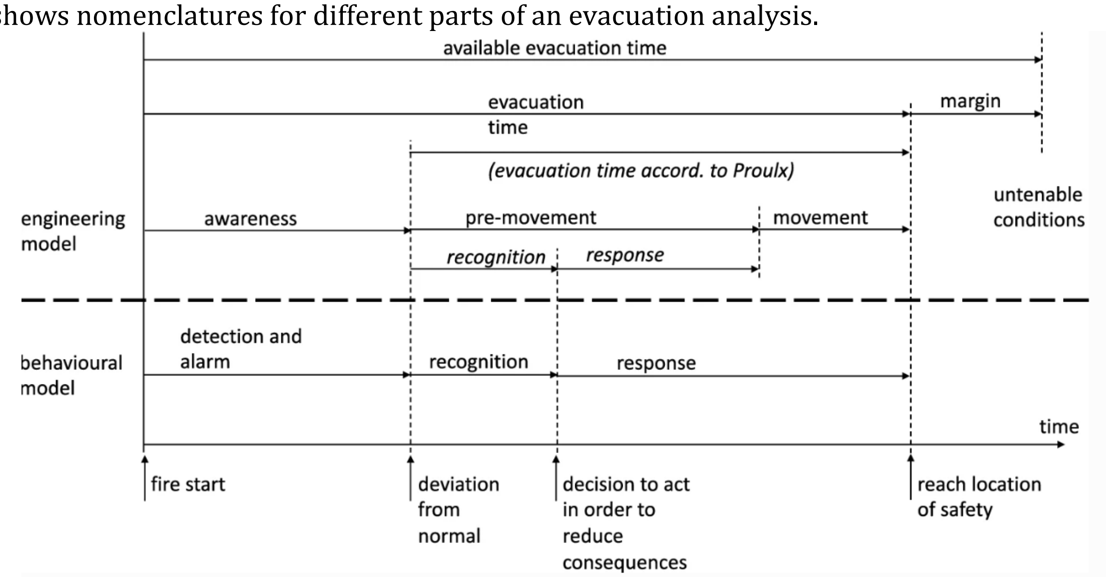

Figure 5. Relation between the engineering model for evacuation analysis and the model to explain human behaviour in fire. [9].

Eight clusters of fire scenarios involving radioactive materials and evacuation scenarios were chosen as representative for TCC2/TDC2. A cluster of fire scenarios is represented by its enveloping credible worst-case scenario. The enveloping credible worst-case scenarios were named CFS1, CFS2, CFS3, CFS5, CFS6, SFS1, SFS2 and SFS3. The results are shown below. Figure 6 demonstrates that the largest portion of the total evacuation time is the detection and alarm time followed by the pre-movement time. Because of potentially long detection times, the worst-case time of about 9 minutes detection and alarm plus pre-movement time after the start of a fire has been chosen to be representative of an enveloping conservative worst-case scenario. If evacuation happens faster than this worst-case scenario, a smaller mass of soot and CO and consequently lower activities would be inhaled.

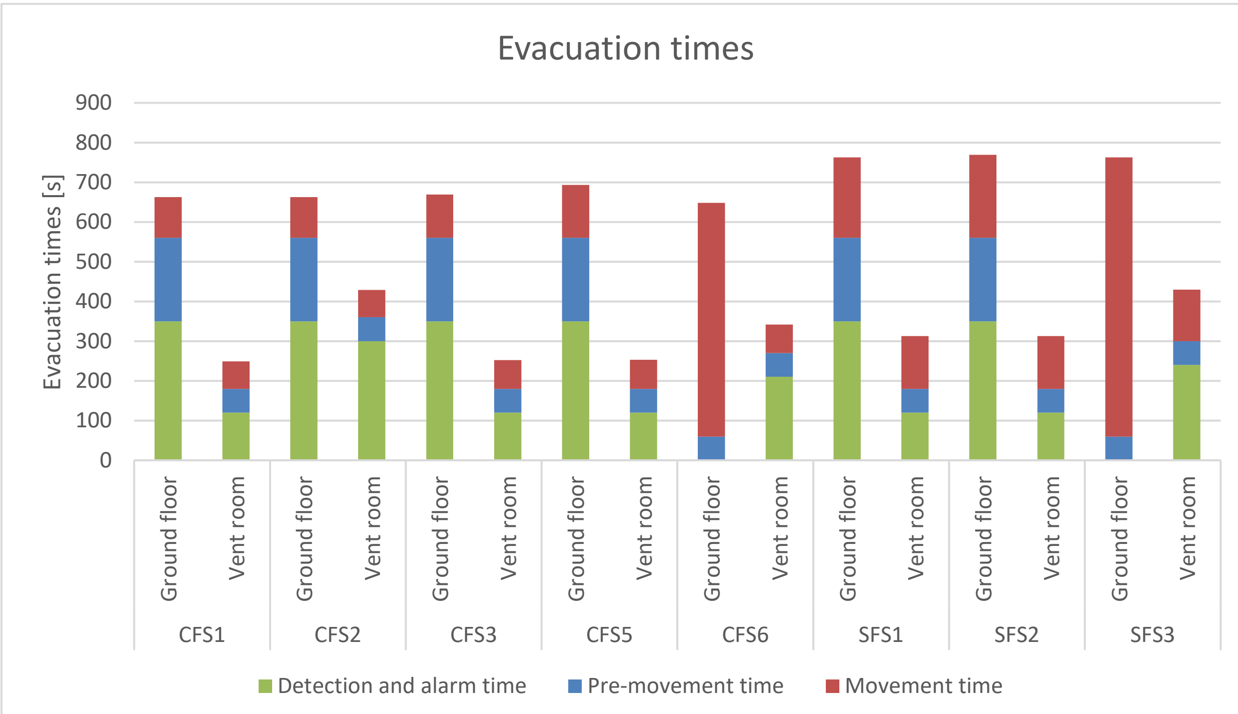

Figure 6. Total evacuation time (last occupant). This was divided into two groups, people present on the ground floor (U0); and people present in the ventilation room at the junction area between TCC2 and TDC2.

It is also dependent on the location and the evacuation direction of the occupant in the facility. Soot and CO concentrations are highest close to the ceiling since hot smoke rises. This is reflected in the inhalation doses depending on the location and evacuation direction of the occupant.

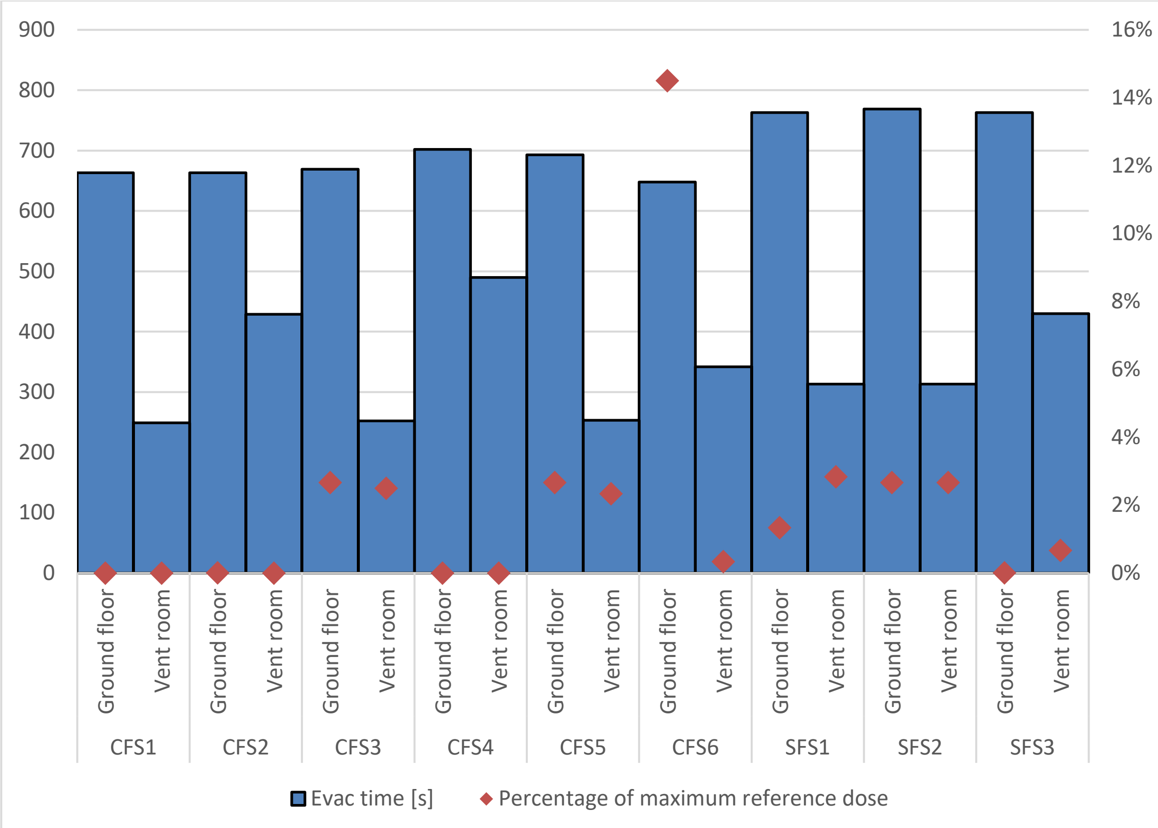

Figure 7. The inhalation dose depends on the location of the occupant in the facility and evacuation time.

The outlying value in CFS6 is related to the specific evacuation scenario. In this worst-case, one of the occupants would be present in the TDC2 area and a fire starts at the end of the TDC2 area, about 50 m from the door towards TT20. The fire would be initiated by a transport vehicle fire spreading to magnet resin and cables in the area. The simulation program chooses the evacuation path for each occupant based on visibility and distance to the exit. For this specific occupant, an evacuation path was chosen through the smoke, past the fire, towards TT20, as shown in Figure 8. The total evacuation time would be 648 seconds. This scenario is highly unlikely but even under such conditions, the received inhalation dose would be lower than 15% of the dose constraint. The main contributors to the inhalation dose are ^{124}Sb, ^{122}Sb, ^{35}S, ^{32}P, and ^{22}Na that account for 99.3% of the total inhalation dose (Figure 9). These radionuclides have long half-lives compared to the typical access delays of several hours, hence, the results are insensitive to this parameter.

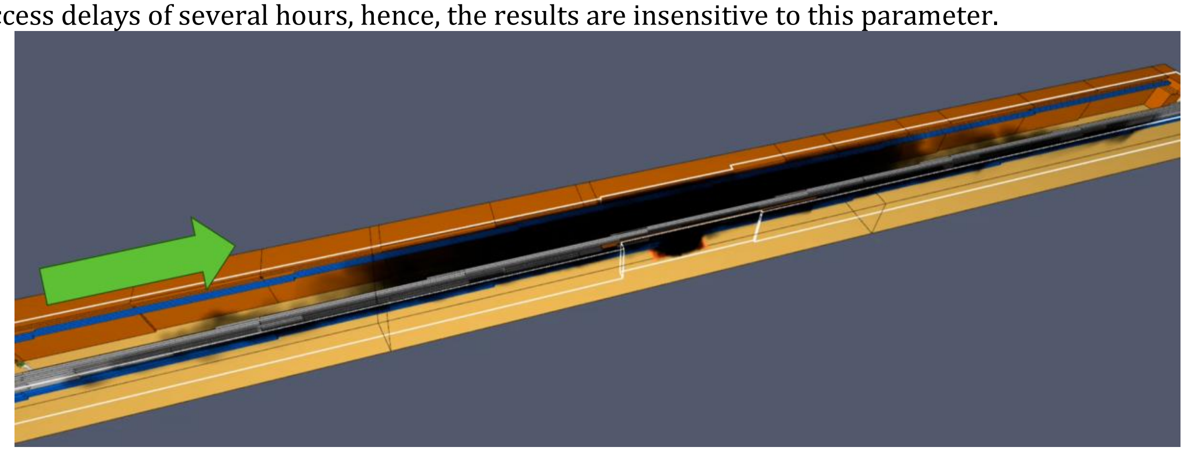

Figure 8. The credible worst-case combined fire and evacuation scenario – an evacuation through smoke towards the nearest exit.

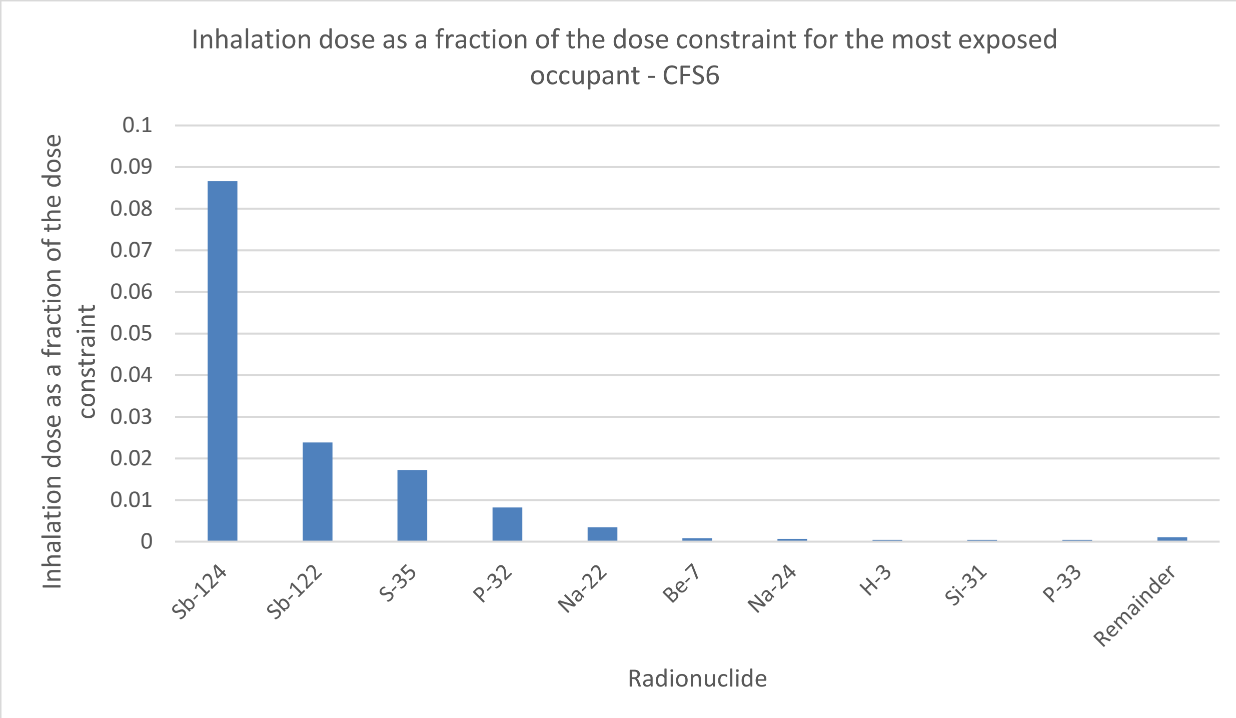

Figure 9. Breakdown of the inhalation dose according to the most important radionuclides for the most exposed occupant in the fire scenario CFS6.

By contrast, the same evacuee using an evacuation direction away from the fire inhaled may receive an inhalation a dose of only 0.003% of the dose constraint. Evacuees who would pass at a longer distance from the fire would receive no inhalation dose because the buoyancy would push the warm smoke upwards to the ceiling of the large and tall hall. However, people working in the CV room in the junction area of TCC2 and TDC2 could receive comparable inhalation doses although evacuation times are shorter for the CV room.

CONCLUSION

The novel methodology allows to quantify the radiological effective dose due to inhalation using Pathfinders moving agents submodel.
It's a highly adaptable methodology for the presence of trace gases, volatile components, liquids or solids, by changing the specific activity to a specific content of a substance. This could for example be used to evaluate the effect of lead content in a combustible, asbestos content, etc.
Using this novel methodology is currently however dependent on post-processing of output provided by Pathfinder and it could be beneficial to add this capability to the software.
In a later stage, if more flexibility exists in the definition and coupling of PLOT3D properties, more properties can be coupled, and consequently this workaround would no longer be needed.

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