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RESEARCH ARTICLE

STUDY OF PARTICULATE AND GASEOUS DEPOSITION (DS) OF HEAVY METALS RELEASED INTO THE SOILS OF THE COMINAK AND SOMAIR MINING AREAS: CASE OF U-238 AND TH-232

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Abstract

The COMINAK and SOMAIR mining sites are among the largest sites in the world for the extractionof uranium (U-238) and thorium (Th-232) metals. The average value of uranium U-238 and thoriumTh-232 concentrations from our measurement results carried out around these two sites are respectively 2.34 ppm and 11.7 ppm in the soils of the COMINAK mining area, compared to 1.35ppm and 7.85 ppm in the soils of the SOMAIR mining area. These average concentration values are below the global normal value for U-238 which is 2.95 ppm (35 Bq/Kg), while they exceed the normal value for Th-232 which is (30 Bq/Kg) or 7.39 ppm in these two sites. To date, the exact value of the particulate and gaseous deposit (Ds) of the annual release of these metals into the soils of the COMINAK and SOMAIR mining areas is not known due to the different parameters. It is therefore necessary to carry out studies on the evolution of the input of these pollutants to the soil. Here, we study in recurrence, the evolution of the input Ds of the metals uranium (U-238) and thorium (Th-232) to the soil by mainly considering that the source of contamination is linked to the extraction of these metals in said sites in order to estimate the content of these metals in the years to come. It emerges from this study that: - For the COMINAK site which is shut down, the thorium concentration regularizes in 29 years if the shutdown is prolonged; - if the extraction of these metals continues in these two sites, the increase in the content of these metals increases if the deposit Ds of the annual discharge of these metals exceeds a limit value discussed in this article according to the site and the nature of the metal considered. And this could have a harmful impact on the environment.

Introduction:-

Uranium mining in Niger began in 1968. Since then, approximately 68,917 tons of uranium has been extracted from the national subsoil.Currently, two companies, SOMAÏR and COMINAK, are mining uranium in the country at a rate of 2,960 tons/year.In 1996, Niger was the world's third largest uranium producer with 3,321 tons produced [11]. Thus, during uranium mining, large quantities of solid waste are generated and stored in the form of slag heaps.These abandoned mining wastes, some of which are undeveloped and subject to climatic hazards, particularly wind and precipitation represent a source of contamination of the environment (water, soil, plants) near these sites by heavy metals.In addition, heavy metals are generally referred to as naturally occurring metallic elements with a

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density greater than 5 g/cm^3 and any metal with a high atomic number, generally greater than or equal to that of titanium (Z=22). Soil not only serves as a medium on which plants grow, but also provides a habitat for living beings and other microorganisms [1].Therefore, soil is a vital part of environmental, ecological and agricultural resources that must be protected for sustainable development and future generations [2].Recent research has shown that soils in different parts of the world, especially in urban and industrial areas, contain extremely high concentrations of heavy metals [3, 4].Work on heavy metals has begun to attract the attention of scientists because of their non-biodegradability, toxicity, persistence, and prevalence [5].Our research revealed that the average values of U-238 and Th-232 concentrations in the soils of the COMINAK mining area are respectively 2.34 ppm and 11.695 ppm compared to 1.345 ppm and 7.85 ppm in the soils of the SOMAIR mining area in the Arlit department [6].Given these measured values, only the thorium concentration for the COMINAK site exceeds the global limit of 7.39 ppm.The COMINAK mine has been shut down for some time, which allows us to monitor the radioactive decay of these metals in the COMINAK soil if the shutdown is prolonged. After the shutdown of an industrial activity, the soil environment is often the main source of exposure.In the case of an operating site emitting atmospheric emissions, exposures (apart from direct inhalation of pollutants) depend to a large extent on the concentrations in the soil environment, which is a major receptacle for environmental contamination.Determining the concentration of pollutants in soils is therefore one of the first steps in the exposure estimation process.From the concentration in soils, the pollutant flows to other environmental media (air, groundwater, surface water) are estimated, as well as the contamination of plants, and the exposure of animals and people by inhalation, soil ingestion and skin contact[12].Currently, the exact value of the particulate and gaseous deposition (Ds) of the annual release of these metals into the soils of the COMINAK and SOMAIR mining areas is not known due to the different parameters. It is therefore necessary to carry out studies on the evolution of the contribution of these pollutants to the soil. Here, we study in recurrence, the evolution of the contribution Ds of the metals uranium (U-238) and thorium (Th-232) to the soil by mainly considering that the source of contamination is linked to the extraction of these metals in said sites in order to estimate the content of these metals in the years to come. This work aims on the one hand to study the evolution of Ds over time knowing the concentration of radioactivity measured in the soils of the COMINAK and SOMAIR mining areas, but also to follow the variation in the concentration of these heavy metals in the soil as a function of time and by fixing the value of Ds.

Methodology:-

In this methodology part, a brief notion on the evolution of a particulate deposit containing a pollutant X as well as the method used was exposed. We study in recurrence, the evolution of the particulate deposit Ds of the metals uranium (U-238) and thorium (Th-232) on the ground by considering mainly that the source of contamination is linked to the extraction of these metals in its sites in order to estimate the content of these metals in the coming years. In this approach, the method of integration of the radionuclide migration equation was treated with the Matlab code version 2022a which is simulation software used for numerical calculation purposes [7]. The standard values of the migration coefficients in the soil are: 1.65E-9s^{-1} for uranium and 2.19E-10 s^{-1} for thorium [8]. The average values of the concentrations of uranium U-238 and thorium Th-232 integrated into the Matlab calculation code are respectively 2.34 ppm and 11.7 ppm in the soils of the COMINAK mining area, compared to 1.35 ppm and 7.85 ppm in the soils of the SOMAIR mining area. The time interval considered is from 0 to 200 years for an iteration step of five years.

Calculation of the concentration of a pollutant X in the soil.

The migration of metals in the soil is the result of the conjunction of several phenomena: dispersion, diffusion, convection, radioactive decay and physicochemical exchanges.

Let Ds (in mg.kg^{-1}.s^{-1}) be the particulate and gaseous deposit of a pollutant containing a metal X in a soil considered; k (s^{-1}) be the loss factor of the pollutant in the soil by the attenuation phenomenon of the metal X and C_0 be the concentration of the metal X in this soil at the initial time t_0=0.

At time t, the concentration of pollutant in the soil is given by the following differential equation [10]:

$$\frac{dC_s(t)}{dt} = -C_s \times k + D_s \quad (1)$$

Where Cs is the Concentration of pollutant in the soil at time t

The general solution of the previous equation is of the form:

$$C_s(t) = C_s(0)e^{-k\times t} + \frac{D_s}{k} \times \left(1 - e^{-k\times t}\right) \quad (2)$$

Let T_1 and T_2 be the exposure times, the average concentration of pollutant X in the soil during the exposure period is calculated as follows:

$$\overline{C_s} = \frac{1}{T_2 - T_1} \int_{T_1}^{T_2} (C_s(0)e^{-k \times t} + \frac{D_s}{k} \times (1 - e^{-k \times t}))dt \quad (3)$$

With ($\overline{C_s}$): Average concentration of pollutant in the soil from T_1 to T_2;
T_1: start of the exposure period;
T_2: end of the exposure period.
If the loss factor of the pollutant in the soil by the phenomenon of attenuation of the metal is zero, the concentration of the pollutant in the soil at time t is reduced to the following expression

$$C_s(t) = D_S \times t \quad (4)$$

And the average concentration over the duration of exposure is expressed by the following equation:

$$\overline{C_S} = \frac{D_s}{2} \times (T_1 + T_2) \quad (5)$$

Expression of particulate deposition DS as a function of concentration:
Heavy metals are naturally present in trace amounts in soils, depending on their geological characteristics (natural pedo-geochemical concentration). Natural fallout processes of volcanic origin, for example, also influence concentrations. Human activities (whether domestic, industrial or agricultural) also influence concentrations by depositing dust on soils and water. Soil erosion contributes to suspending metals in particulate or gaseous form.

According to equation 2, the deposition Ds is linked to the concentration C by the following formula:

$$Ds = k \frac{C - C_0 e^{-kT}}{1 - e^{-kT}} \quad (6)$$

With k the loss factor of the pollutant in the soil (in years minus one);
T is the exposure time in years;
C_0 the initial concentration of the metal
And C the concentration at time T

Assuming that the concentration of a heavy metal follows the geometric progression of reason q whose recurrence expression is of the form: $C = C_0.q^T$ the expression of the deposit Ds becomes:

$$D_s = kC_0 \frac{q^T - e^{-kT}}{1 - e^{-kT}} \quad (7)$$

Then assuming that the concentration follows the arithmetic progression of reason r, whose recurrence formula is: $C = C_0 + T \times r$, the expression of Ds becomes:

$$D_s = k. \frac{T \times r + C_0 \left(1 - e^{-kT}\right)}{1 - e^{-kT}} \quad (8)$$

Exposure dose related to soil ingestion
Exposure doses are determined substance by substance; it is not possible to determine exposure to several substances at the same time [9]. For the same substance, exposure doses via the different routes taken into account in the use of the site are presented, then summed (regardless of the oral, respiratory or cutaneous administration method) while the effects depending on the different routes considered may be different.

The exposure dose Es related to soil ingestion is given by [10]:

$$E_s = \frac{Q_S \times B_s \times C_s}{P} \times \frac{F}{365} \quad (9)$$

With Qs: Quantity of soil ingested per day per person (kg/days);
Cs: Concentration of pollutant in the soil (ppm);

Bs: Relative bioavailability factor of the pollutant in soil compared to the bioavailability of the pollutant in plants;

F: Number of days of exposure per year;
P: Weight of the target (Kg).

Results and Discussion:-

Table 1:- Forecast of the Ds of U-238 in soil assuming that the concentration follows a geometric and arithmetic progressions of reason q and r respectively and for some values of q and r.

reason	sites	10	20	30	40	50	Times in years
q=1.001	COMINAK	0.124791	0.1256	0.1265	0.1275	0.12851	Ds in ppm.year^{-1}
	SOMAIR	0.0717282	0.0722	0.0727	0.0733	0.07387	
q=1.05	COMINAK	0.22552	0.2741	0.3423	0.4371	0.56742	
	SOMAIR	0.129624	0.1576	0.1968	0.25123	0.326143	
r=0.01	COMINAK	0.134653	0.1379	0.1416	0.14556	0.1499	
	SOMAIR	0.08287	0.086	0.0898	0.0938	0.098113	
r=0.5	COMINAK	0.766399	0.9294	1.0607	1.3132	1.5288	
	SOMAIR	0.7664	0.8776	1.0607	1.261	1.4770	

Table 2:-Forecast of the Ds of Th-232 in soil assuming that the concentration follows a geometric and arithmetic progressions of reason q and r respectively and for some values of q and r.

reason	sites	10	20	30	40	50	Times in years
q=1.001	COMINAK	0.08310	0.083534	0.08398	0.08423	0.08489	Ds in ppm.year^{-1}
	SOMAIR	0.05578	0.0561	0.056366	0.05667	0.05698	
q=1.05	COMINAK	0.83393	1.1041	1.4961	2.062	2.9256	
	SOMAIR	0.5598	0.7412	1.0042	1.3905	1.9638	
r=0.01	COMINAK	0.0813	0.0816	0.08192	0.08225	0.08256	
	SOMAIR	0.05798	0.05829	0.05861	0.05893	0.059261	
r=0.5	COMINAK	0.5896	0.6052	0.6212	0.6374	0.6539	
	SOMAIR	0.5663	0.58193	0.59785	0.6142	0.6306	

Tables 1 and 2 give an estimate of the particulate deposit Ds according to years by assuming respectively that the annual variation of the concentration follows a geometric progression of reason q, then arithmetic progression of reason r for the COMINAK and SOMAIR sites. For these two cases of regressions, we naturally note that Ds increases with the years and according to the reasons of the regressions. The COMINAK site is shut down so our forecast would be on the SOMAIR site. Therefore for the SOMAIR site, the Uranium concentrations which follow the geometric progression of reason q=1.05 for a ten-year and fifty-year forecast are respectively 2.20ppm and 15.48ppm. In ten years the concentration is below the normal world value which is 2.95ppm (35 Bq/Kg) while it is 5 times higher than the normal value for a fifty-year forecast. The Uranium concentrations that follow the arithmetic progression of reason r = 0.05 for a ten-year and fifty-year forecast are respectively 1.4ppm and 3.85ppm. In ten years the concentration is below the global normal value while it exceeds the normal value for a fifty-year forecast. These high concentration values for a 50-year forecast are alarming because there is a risk of contamination of plants by the mechanism of transfer of pollutants from the soil to the plants, of exposure of animals and people by inhalation, ingestion of soil and skin contact [12]. For thorium the average concentration measured on the two sites exceeds the global normal value which is 30Bq / Kg or 7.39ppm. For the COMINAK site which is shut down the thorium concentration will regularize in 29 years if the shutdown is prolonged.

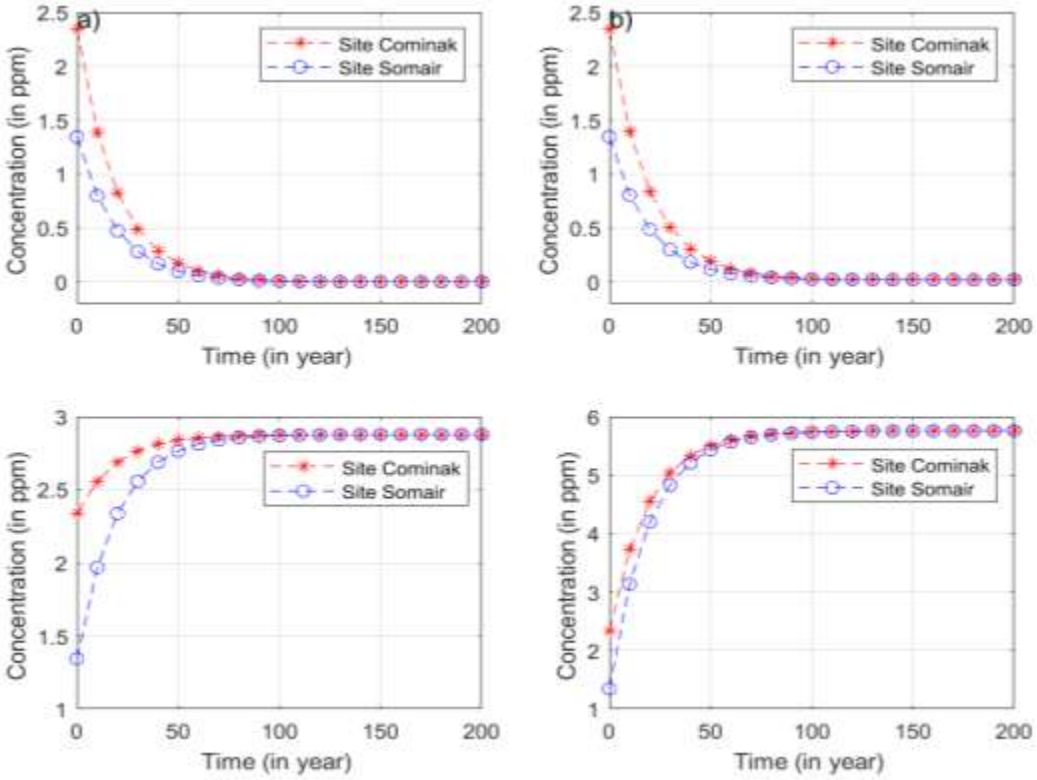

Fig1:- Variation of U-238 concentration in soil: a) Particulate deposition Ds=0.00, b) Ds=0.001, c) Ds=0.15, d) Ds=0.3.

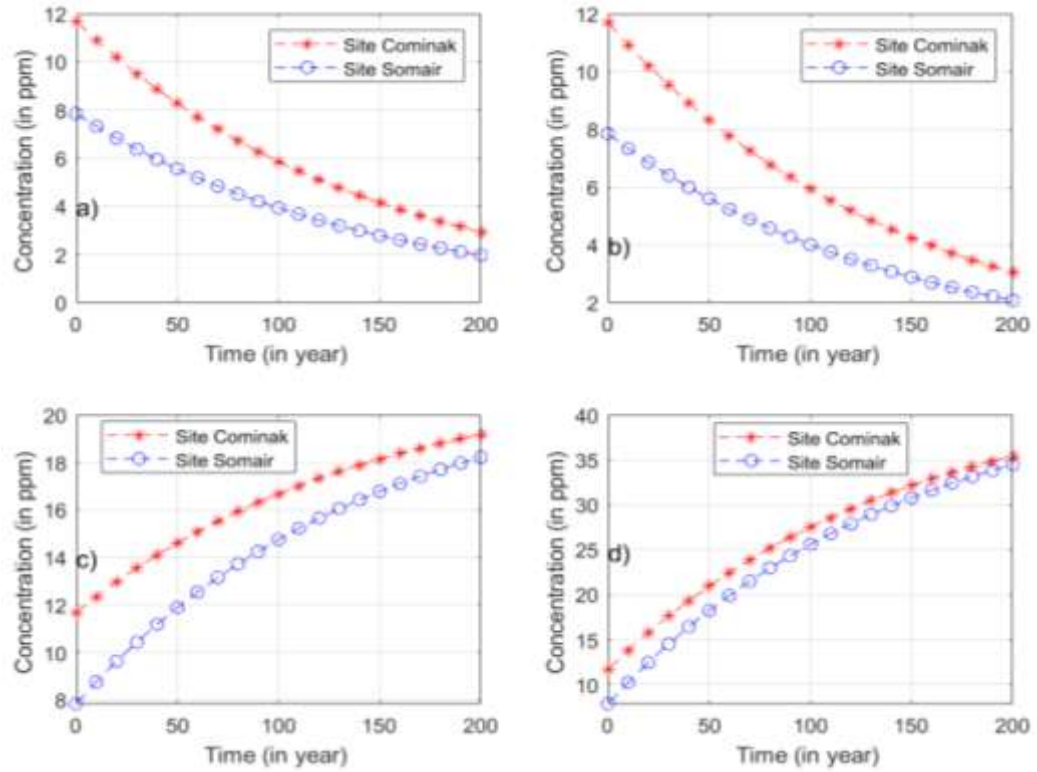

Figure 2:- Variation of Th-232 concentration in soil: a) Particulate deposition Ds=0.00, b) Ds=0.001, c) Ds=0.15, d) Ds=0.3.

Table 3:- Forecast of concentration according to particulate deposition of the metal.

sites	metals	0.04	0.05	0.06	0.07	0.08	0.09	0.1	0.11	0.12	0.13	0.14	D_S
COMINAK	U-238	---	--	--	--	--	--	--	2.19	2.32	2.44	2.57	Ci in ppm
	Th-232	10.93	11.12	11.31	11.49	11.68	11.87	12.05	--	--	--	--	
SOMAIR	U-238	---	--	1.21	1.35	1.47	1.59	--	--	--	--	--	
	Th-232	7.58	7.78	7.96	8.14	-----	----	--	--	--	--	--	

Figures 1 and 2 show the variation of the concentration of the metal Uranium 238 (U-238) and Thorium (Th-232) in the soil as a function of time and for some values of the particulate deposit Ds of the pollutant released into the soil. The curves in round dots give the results of the concentration of the metal uranium in the soil of the COMINAK zone and the curves in starred dots are the results of the concentration of the metal uranium in the soil of the SOMAIR zone. By observing these curves, we see that for a very low value of the particulate deposit Ds of the pollutant in the soil Fig.3,a), Fig.3,b) and Fig.4,a), Fig. 4,b), the concentration of the pollutant in the soil decreases over the years. At Ds = 0.00 the concentration of the pollutant in the soil follows a radioactive decay law with a half-life period ln2/lamda. In 10 years the concentration of Uranium metal in the COMINAK and SOMAIR sites takes the value 1.39015ppm and 0.79904ppm respectively it decreases with time until it reaches saturation in 100 years 0.0074ppm for both sites. That of Thorium metal in the COMINAK and SOMAIR sites is 10.914 ppm and 7.326ppm respectively in 10 years, they would be in a century 5.859ppm and 3.933ppm. This reduction in the concentration of pollutant in the soil to Ds = 0.00 over time, can be linked by the phenomena of leaching, erosion, runoff, volatilization or degradation. And for considerable values of the particulate deposit Ds of the pollutant in the soil, Fig.3, c), Fig.3, d) and Fig.4, c), Fig.4, d), the concentration increases rapidly over the years. For Ds = 0.3ppm.year $^{-1}$ the concentration of uranium in the COMINAK and SOMAIR site at 10 years is 3.73ppm and 3.14ppm respectively, it increases over the years to reach a saturation of 5.74ppm in 100 years. This saturation value of the concentration exceeds the normal value which is 2.97ppm. For Thorium the concentration is of the order of 13.8127 ppm and 10.2244ppm in 10 years respectively for COMINAK and SOMAIR it increases considerably over time to reach 27.52ppm and 25.59ppm respectively in a century. The highest average concentration for the elements analyzed is obtained at the COMINAK site. The analysis of these results clearly reveals that the COMINAK mining soils are richer in U-238 and Th-232 than the soils of the SOMAIR mining area. This is partly explained by the fact that the extraction in the SOMAIR site is done in open pit unlike that of the COMINAK site.

Table 3 provides a forecast of the uranium and thorium concentration in order to determine the limit value of Ds for which the concentration of the pollutant increases. For the COMINAK site, this limit value of Ds is of the order of 0.125 ppm/year for uranium and 0.085 ppm/year for thorium. For the SOMAIR site these values are respectively 0.075 ppm/year and 0.055 ppm/year for the metal Uranium and thorium. This means that below the limit value of Ds, the concentration of metals decreases over the years while this concentration increases beyond the limit value of Ds.

Conclusion:-

During this study, an investigation on the evolution of Ds over time was carried out. A simulation was carried out using the Matlab tool. Thus, this simulation allowed us to study the variation in the concentration of U-238 and Th-232 metals according to years and by fixing some characteristic values of the particulate deposition of the pollutant in the soil. It emerged from this study that the COMINAK mining soils are richer in U-238 and Th232 than the soils of the SOMAIR mining area. This is partly explained by the fact that extraction in the SOMAIR site is done in the open air unlike that of the COMINAK site. The COMINAK site is closed so our prediction would be on the SOMAIR site. For a ten-year and fifty-year forecast, the Uranium concentrations for the SOMAIR site are respectively 2.20ppm and 15.48ppm assuming that the evolution of the concentration follows a geometric progression with a reason q=1.05. The concentration value for a fifty-year forecast exceeds 5 times the global normal value which is 2.95ppm (35Bq/Kg). Similarly, assuming that the Uranium concentration follows the arithmetic progression with a reason r=0.05 for a fifty-year forecast, the value obtained exceeds the normal value. These high concentration values for a 50-year forecast are alarming because there is a risk of contamination of plants by the mechanism of transfer of pollutants from the soil to the plants, of exposure of animals and people by inhalation, ingestion of soil and skin contact. For thorium, the average concentration measured on the two sites

exceeds the global normal value which is 30Bq/Kg or 7.39ppm. For the COMINAK site which is shut down, the thorium concentration will stabilize in 29 years if the shutdown continues.

The approximate value of the Ds deposit could be determined by taking radionuclide measurements for a certain number of years. It should be noted that the health impacts are different depending on the chemical form in which the metal is found in the environment. The impact depends on their concentrations, their bioavailability and their ability to enter the food chain. The results of this study will allow us to understand the path of a possible transfer of metals from the soil to the water and to the plants in order to determine the radiological impact on humans through ingestion of water, of plants having ingested this water or having consumed contaminated plants, which is another project currently being finalized.

This is why, in perspective, we recommend carrying out annual and repetitive sampling of these sites in order to have an estimate of the annual release of these heavy metals in the SOMAIR mining area which is still in operation.

References:

[1]. Sabo A., Gani A.M., Ibrahima A.Q., J. Environ. Pollut.and Human Health 2 (2014) 91-9
[2]. Ameh E.G., Res. J. of Env. and Earth Sc. 6 (2) (2014) 57-65.
[3]. Wei B., Yang L., Microchem. J. 94 (2010) 99–107.
[4]. Duong T.T.T., Lee B.K., J. Environ. Manag. 92 (2011) 554–562.
[5]. Nasr S.M., Okbah M.A., Kasem S.M., IJOO 1 (2006) 99−109.
[6]. Yacouba S.A et al (2024). A comparative analysis of radioactive material concentrations in natural sources linked to mining in Niger using two measurement methods.
[7].The MathWorks, Inc. 1 Apple Hill Drive Natick, MA 01760-2098 MathWorks Installation Help © COPYRIGHT 1996–2022 by The MathWorks, Inc
[8]. DQSSE- Management QSSE Rapport identifiant Framatome/ Framatome Id: ENV-20-005
[9].The MathWorks, Inc. 1 Apple Hill Drive Natick, MA 01760-2098 MathWorks Installation Help © COPYRIGHT 1996–2022 by The MathWorks, Inc
[10]. INERIS (Institut National de l'Environnement Industriel et des Risques), Bonnard, R., les modèles multimédia pour l'évaluation des expositions liées aux émissions atmosphériques des installations classées, DRC-01-255584-ERSA-RBn-n°445, 2001
[11]. Rapport établi conjointement par l'AEN et l'AIEA titré ''Aspects environnementaux de la production d'uranium''
[12].www.ineris.fr.