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Calculation methods of Radon-222 radiological activity for NORM plant with ventilation



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ABSTRACT

A procedure is proposed to determine the radiological activity of air present in the working area of a NORM (Naturally-Occurring Radioactive Materials) plant. This NORM plant is located in the United Arab Emirates and is dedicated to the recycling of material and equipment used in oil extraction facilities. Substantial quantities of NORM waste will be present inside the enclosed plant facilities and will exhale significant amounts of Radon-222 into the working environment. The continued inhalation of this gas and its progeny has been shown to cause lung cancer. In order to reduce the concentration level of the aforementioned gas to an acceptable regulatory value, the best solution is by means of mechanical ventilation. The two calculation methods used to analysis the relationship between the ventilation rate and the degree of radioactive contamination are considered. The first being ventilation by perfect dilution, commonly employed in industrial environments with radiological contamination. The second method of analysis is by means of a CFD (Computational Fluid Dynamics) which permits a more precise calculation of the required fresh air quantity and spatial concentrations. Validation was carried out in a small experimental plant space, obtaining results reasonable approximate to those predicted by the computational analysis. The similarities and differences between the respective calculation methods and their respective fields of application are analyzed.

It is concluded that for lower air change rates, less than five air changes per hour, the CFD analysis is the most appropriate method. By contrast, when using a higher rate of outside air changes in the plant, the perfect dilution method should be sufficient enough to control the presence of radon and its progeny. Both calculation methods consider the activity of radon and the activity of the most significant radioisotopes of its progeny.

1. Introduction

All living beings are exposed to ionizing radiation. Most of this radiation is of natural origin. An important source of radiation from radioisotopes breakups of natural radioactive chains are ^{238}U and ^{235}U uranium and thorium ^{232}Th (Arafa, 2004; Rahman et al., 2010; Agarwal et al., 2014; Ghany et al., 2018). In general, many rocks contain natural radiation, as is the case with sludge and rock sediments from oil wells, which include all of the mentioned radioisotopes decay chains (Taylor and McLennan, 1985; Kovler et al., 2002; Sakoda et al., 2011). These three chains contain a single gaseous element, radon, each chain having a different isotope, ^{222}Rn , ^{219}Rn and ^{220}Rn , with half-lives of 3.8235 days, 3.9 s and 55.6 s, respectively (Lara et al., 2015). The isotope ^{222}Rn

is the only one which has a half-life long enough to permeate through the NORM waste into air (El-Dine et al., 2001; Ferrero, 2013).

^{238}U , uranium, precursor of ^{222}Rn , is widespread in the earth. In addition, the half-life of ^{238}U is very long (of the order of 4500 million years), reason for why the formation of Radon will continue to happen for tens of millions of years almost in the same concentration that occurs today (Righi and Bruzzi, 2006). ^{222}Rn inhalation leads to radioactivity in the lung. Its progeny, the short-lived radioisotopes derived from its disintegration (^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po), are a radioactive focus in the lung. Even the stable dust containing lead at the end of the chain, ^{206}Pb , is highly toxic although nonradioactive. The pathology associated with radon is lung cancer (Chen et al., 2010). Epidemiological studies have shown a clear link between breathing high

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Nomenclature

Ac^{Xy}	Activity of 1 m ³ of contaminated air due radioisotope Xy, Bq·m ⁻³
M^{Xy}	Molecular mass of radioisotope Xy, g·mol ⁻¹
m^{Xy}	Mass of radioisotope Xy present in 1 m ³ of contaminated air, g
N_{av}	Avogadro's number, 0.60225·10 ²⁴ atoms·mol ⁻¹
N^{Xy}	Number of atoms of radioisotope Xy in 1 m ³ of

	contaminated air
Q	²²² Rn escaped from NORM waste to the indoor air, 1.93·10 ¹² atoms·h ⁻¹
R	ventilation air change rate in the plant, h ⁻¹ and s ⁻¹
T^{Xy}	Half-life of radioisotope Xy, time units (y, d, m, s)
t	Time, s
Vr	Volume of polluted air in the industrial plant, m ³
λ^{Xy}	Decay constant of radioisotope Xy, s ⁻¹

concentrations of radon and the incidence of lung cancer (Castren et al., 1985). Therefore, radon is considered a key contaminant that seriously affects indoor air quality when found in high concentrations in occupied spaces. According to the Environmental Protection Agency of the United States, radon is the number one cause of lung cancer among non-smokers (ATSDR, 1990; EPA, 2003). This serious associated pathology coupled with the fact that it is very difficult to detect (it is colorless, odorless and tasteless), and that as it has no chemical reactivity (a noble gas) makes Radon an extremely dangerous gas (Higgy et al., 2000; El-Hussein, 2005).

In the plant case study, sludge impregnated with hydrocarbons is removed from the drilling equipment, where this sludge has a high content of ²²⁶Ra that exhales ²²²Rn. The subsequent disintegrations of the ²²²Rn chain gives rise to the dangerous progenies that will adhere by electrostatic charge to the dust particles existing in any industrial plant. This progeny suspended in the air is a serious long-term health hazard for the people exposed to it, in any working or living environment, increasing considerably the chances that they develop lung cancer (Chao et al., 1997a,b; Wang and Ward, 2002; Stoulos et al., 2003).

In the field of occupational safety, there are guidelines regarding radon concentrations (EC, 2001; IAEA, 2006; WHO, 2009; Chauhan et al., 2014). Mandatory regulations also exist both internationally (UNSCEAR, 2006; WHO, 2007; ICRP, 2009), in Europe (EC, 1997, EC, 1999a,b) and on a national level in Spain (BOE, 2010). There is also information regarding measurement procedures of radon concentration (Tung et al., 2005). Because of the null chemical activity of radon, the only way to reduce its concentration is by means of ventilation (Cavallo et al., 1996, Chao and Hu, 2004).

The majority of existing case studies related to radiological contamination, due to radon, have been carried out in and for homes and their focus being on single-family homes (Akbari et al., 2013). The analytic consequences in the before mentioned studies for housing are not applicable to industrial building type applications (IAEA, 2003; Dieguez-Elizondo et al., 2017), and more so in the case of the NORM plants which are usually located in areas where the air from the environment, to be used in supply air the ventilation system, contains a considerably higher concentration of radon due the NORM waste disposal areas and refinery equipment storage which are present. This outside radon concentration is not considered here in this study given that the object of this study is to provide a reliable method analysis approach, it can be derived in further studies the impact of the concentration of radon on the required supply air ratio by measuring the actual value in each application studies. However it can be deduced that the only effect will be an increased supply of the required airflow ventilation rate.

In this research, the radioactivity of ²²²Rn and its progenies in the indoor plant environment is analyzed and what will potentially be inhaled by workers in the breathing zones. It does not form part of this investigation the analysis of the thermal conditioning of the plant or the removal of other pollutants that may exist. The aim of the study is to look at the radon and its progeny/air mixture, and what level of mixture will impact on the radioactivity of air inside the factory. Two hypotheses are considered: the first is the perfect mixing-dilution with a

zero gradient concentration of radon (perfect dilution of radon air) and the second hypothesis is the opposite, considering radon concentration gradients. The first hypothesis is relatively easy to calculate while the second hypothesis cannot be achieved without the use of a CFD analysis given the multiple variables associated.

2. Material and method of analysis

The object of the research is to study the concentration of radon, emitted by the NORM waste, and its progeny.

2.1. The factory layout

The factory consists of several buildings, each of them dedicated to one or several industrial processes, giving rise to a complex and intricate layout. In one of these buildings, the Incinerator plant, the NORM waste is incinerated, passing the ashes to another building, the Solidification plant. In this last building, the ashes are stored in a large Scale Silo and then cemented and stored in Curing Drums waiting for their transport. The Solidification building is considered in this study. This building is compartmentalized in different areas but its morphology and content are been simplified to the maximum in this work. The position of the two sources of radon considered, Scale Silo and Curing Drums are determined by the design considerations of plant layout and process, in the case of Scale Silo distribution logistics, and in the case of Curing Drums they are located in a corner of the plant accessible to the outside for their transport.

Inside the selected plant area is the treated sludge that contains an appreciable concentration of ²²⁶Ra that exhales ²²²Rn. Although the ventilation system of the plant is designed to mitigate the important thermal loads, in this study the objective of the ventilation system is to reduce the concentration of ²²²Rn and its progeny.

The plant area considered has a rectangular dimension of 30.6 × 17.5 m and 16 m in height. In one of the shorter sides, there is a door with a dimension of 5 × 5 m (25 m²). In the upper part of the plant, at the roof level, there is a longitudinal ventilation extract exhaust opening of dimensions 30.6 × 0.8 m (32.48 m²). Inside the plant there are two radon emission sources, the Curing Drums and Scale Silo. The ventilation is accomplished by introducing forced outside air through seven conduit inlets each of 1 m², and equally spaced along the length of the wall and at a height of 2 m from the floor to their inferior part; the extraction is achieved through the roof opening (Fig. 1).

For obtaining the radon exhalation rate the approach recommended in the IAEA Technical Report 474 (IAEA, 2013) has been used. In the plant case study, there are two sources of radon from the NORM handling process. The most important is Curing Drums consisting of 160 cement drums (ashes) with a content of ²²⁶Ra equivalent to 917 Bq·cm⁻³. The other major source of emission is the Scale Silo, which is the dust collection system, and has a ²²⁶Ra content equivalent to 1288 Bq·cm⁻³. The emanation values are generally lying in the range of 0.1–0.3, where 0.2 has been assumed for cemented ashes (medium size particles and 80% water saturation assumed) and 0.15 for Scale (big size particles and same water saturation). Radon diffusion coefficients are generally dependent on the waste type, its pore size distribution, its

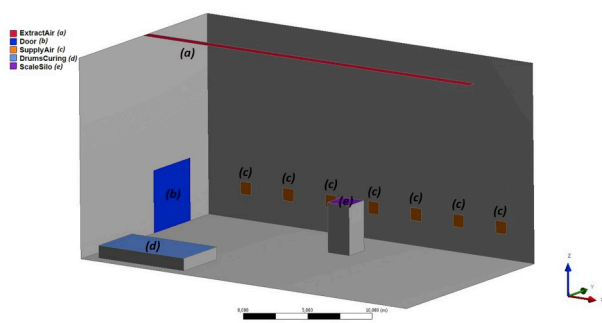


Fig. 1. Factory layout, radon emission sources and ventilation system.

water content, and the degree of its compaction. The diffusion coefficient value has been assumed the following way, with an equivalent to a soil with 80% of water saturation that is $10^{-7} \text{ m}^2 \text{ s}^{-1}$ and multiplying the flux density by the waste package surface (residue matrix-air interface) the radon exhalation rate is obtained. Thus, the exhalation of ^{222}Rn atoms leaving both waste matrix is $1.30 \cdot 10^{12} \text{ }^{222}\text{Rn atoms h}^{-1}$ for Curing Drums and $6.30 \cdot 10^{11} \text{ }^{222}\text{Rn atoms h}^{-1}$ for Scale Silo, that is, the exhalation is in the order of $2 \cdot 10^{-13} \text{ g s}^{-1}$ for the plant area referred to in this study.

Radon emissions from the building walls, floor or other construction material are not considered, nor the radon introduced from outside air by renovation with fresh air.

2.2. The radon-air mixture

Despite being a very small amount exhaled, the radiological impact on occupational health is decisive. In effect, and considering the expression of the radioactive activity (Eq. (1)):

$$Ac = \lambda \cdot (m/M) \cdot N_{av} \quad (1)$$

For a permitted radioactive contaminated air concentration of 400 Bq m^{-3} , considered as a threshold value by most health authorities, as also in the case of the plant authorities of the NORM handling plant in this study, the amount of radon present in the air, $7 \cdot 10^{-14} \text{ g in } 1 \text{ m}^3$. A single atom of radon per approximately $5 \cdot 10^{17}$ atoms of oxygen-nitrogen makes this air dangerous to health if breathed continuously.

From a chemical point of view and as mentioned before, radon is undetectable by odor or color. As a noble gas, it has a zero chemical reactivity, so chemical based capture techniques or combinations with chemical capture reagents are non-viable. The only presently known and feasible technique for decontaminating the air is by dilution, i.e. the supply and exhaust ventilation technique (Chao et al., 1997a,b; NCRP, 1997).

Such low concentrations are not easily detectable by sensors. Radon measurements are not instantaneous but require a relatively long period of integration (various days or inclusive weeks). This circumstance requires prevention, since it is not impossible to know instantly the radiation exposure due to the radon owing to inhalation by the occupants. Likewise, it is difficult and very time consuming for the experimental determination of a radon map in any given contaminated space.

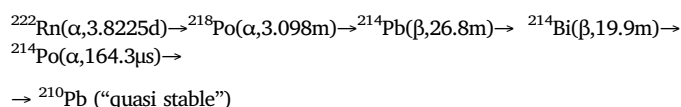
Concerning the interactions of a radon-air mixture, such a small amount of radon present as a specific concentration implicates extreme dilution and therefore do not produce, in practice, density variations, consequently the buoyancy forces are negligible, even though the density of radon is almost eight times that of air. Due to mechanical ventilation, the inertial forces are the dominant forces that will determine the composition of the mixture and therefore, the specific concentrations of radon in the enclosure and its impact on occupancy health.

Radon and air mixing is, as far as its chemical and physical properties are concerned, a gas with so low a percentage of radon that it

does not affect either its physical or its chemical properties, while it certainly does with respect to its radiological properties. Thus, a CFD study of a radon-air mixing is essentially a CFD study of air, in which a small amount of atoms of radon and its progeny is spread through the air like any other atom of the air.

2.3. The progeny of radon

The natural disintegration chain of ^{238}U is well known. In the present work, from the existing ^{226}Ra in the NORM waste ^{222}Rn is generated that permeates through the sludge waste and passes into the air of the factory plant. Considering the radioactive chain of disintegration from ^{222}Rn , two simplifying hypotheses are made. On the one hand, the disintegration of ^{218}Po (β) and ^{214}Bi (α) are obviated since the frequency of both disintegrations is 0.02% and 0.021% respectively. On the other hand, since the average life of ^{210}Pb is 22.2 years, it can be considered “quasi stable” for the purpose of calculating radiological activity in the factory plant. Thus, a simplified radioactive chain present in the air of the factory plant is the following:



This chain is called the progeny of ^{222}Rn , short-lived radioactive decay products of ^{222}Rn that turn into longer-living lead isotopes ^{210}Pb upon decaying, such as ^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po . Unlike the gaseous radon itself, radon progenies are solids and stick to other solids and surfaces due to their electrostatic charge, such as dust particles in the air. If such contaminated dust is inhaled, these progeny particulates are deposited on the lung tissue, where the alpha radiation can damage the cells, increasing the risk of developing lung cancer.

The ^{218}Po and ^{214}Po radioisotopes are especially dangerous: both are high energy alpha particles, 6.11468 and 7.83346 MeV, respectively. If a ^{218}Po atom is deposited in lung tissue, nearby cells will have to support two alpha particles and two beta particles; if the deposited atom were ^{214}Po , the radiation supported would be half. Also note the high toxicity of all progeny, especially Polonium.

The radon progeny (also known as radon daughters) are considered responsible for most of the damaging biological effects, not radon itself. The radon equilibrium factor is the ratio between the activity of all short-period radon progeny, and the activity that would be at equilibrium with the radon parent. This factor is lowered by air recirculation or air filtration devices and is increased by airborne dust particles, and then it is very difficult to discern it. The equilibrium factor found in epidemiological studies is usually 0.4 (IAEA, 2006; ICRP, 2009).

In the present work it is considered that the disappearance of the progeny takes place by air renewal and by radioactive decay. The phenomenon of electrostatic adhesion of progeny to dust, aerosols or walls is considerable and tends to decrease its presence in the air. As this phenomenon of electrostatic adhesion is not considered, the radiological activities of the progeny calculated in the present work will be greater than the real ones, i.e., the calculations presented are conservative. The progeny are considered to be entrained by air, following the airflow patterns, without any interaction with other atoms, particles or bodies in the air.

The phenomenon of depositing on the surfaces or “plate out” of the progeny of radon depends on a large number of factors (morphology of the enclosure, its surface/volume ratio, concentration of aerosols, humidity and temperature, etc.), all of which entails a great complexity, obliging complex experimental studies. However, several such studies (Lagarde et al., 2002) estimate that the concentration of the progeny on the surfaces is approximately the same as that in air, which means that half of the calculated radiological activity will be present in the air. The plate out phenomenon implies that the concentration of radioisotopes in the air decreases at the expense of adhering to the surrounding

surfaces. Considering that the radiological risk is in the air pollution and its subsequent inhalation, not considering the plate out effect implies a calculated concentration of radioisotopes in air greater than the actual air quantity concentration. The non-consideration of the plate out effect is a safety factor.

2.4. The study hypothesis

The consideration of perfect dilution of radon in the air of the plant, or to the contrary, the consideration of the existence of gradients of radon concentration is a determining factor in the mathematical complexity of the problem and, therefore, governs the use of one or the other calculation methods.

In the case of perfect dilution, the concentration of radon is uniform throughout the plant. The phenomenon is governed by a simple differential equation of a single independent variable, time, which is easily integrated.

In cases where ventilation systems work with low velocity, such as is the case with displacement ventilation systems, the perfect dilution method is not applicable (Gil-Lopez et al., 2017). In particular, in displacement ventilation applications, characterized by low turbulent airflow, lower discharge velocities and admixing produce thermal gradients due to the buoyancy effect of the heat source (ASHRAE, 2017). The perfect dilution method is not applicable and a CFD calculation is therefore necessary in order to determine the spatial concentration.

When considering spatial gradients, i.e. the variation of the concentration of radon with respect to any given position, it is fundamental to resolve simultaneously the equations of the conservation of mass, momentum and diffusion, which make it imperative the use of a computational simulation program. The reliability of these calculation programs, in their application to the calculation of radioactive atmospheres, has been demonstrated (Andersen, 2001, de With and de Jong, 2011, Agarwal et al., 2016, Dieguez-Elizondo et al., 2017, Rabi and Oufni, 2017). In this research, the ANSYS CFX V18.1 software has been used.

The following two cases scenarios are analyzed and presented:

Case 1. a case study with ideally uniform, perfect dilution or zero spatial concentration gradients. The well-known theoretical method of study is presented for comparison purposes.

Case 2. consideration of spatial gradients, simulations are presented with forced air for ventilation flow rates denominated R and shown in section 2.4.2. An air tight room operation is assumed, the entire flow enters through the seven lateral openings by means of the mechanical ventilation system where the same airflow rate is extracted through the opening in the ceiling.

2.4.1. Case 1: uniform spatial concentrations

The emission of ^{222}Rn atoms into the air is constant. The elimination of ^{222}Rn atoms depends on two factors: its radioactive decay and ventilation or air change rate, R . The change of rate of ^{222}Rn atoms, with time, are determined by Eq. (2.1).

$$dN^{Rn222}/dt = [Q/V_r] - (\lambda^{Rn222} + R)N^{Rn222} \quad (2.1)$$

The remaining radioisotopes of the simplified chain are generated by disintegration of the above radioisotope and are eliminated (Eqs. (2.1)–(2.6)), similarly to ^{222}Rn by ventilation-renewal with fresh air and by radioactive decay (except the isotope considered “quasi-stable” ^{210}Pb).

$$dN^{Po218}/dt = \lambda^{Rn222}N^{Ra222} - (\lambda^{Po218} + R)N^{Po218} \quad (2.2)$$

$$dN^{Pb214}/dt = \lambda^{Po218}N^{Po218} - (\lambda^{Pb214} + R)N^{Pb214} \quad (2.3)$$

$$dN^{Bi214}/dt = \lambda^{Pb214}N^{Pb214} - (\lambda^{Bi214} + R)N^{Bi214} \quad (2.4)$$

$$dN^{Po214}/dt = \lambda^{Bi214}N^{Bi214} - (\lambda^{Po214} + R)N^{Po214} \quad (2.5)$$

$$dN^{Pb210}/dt = \lambda^{Po214}N^{Po214} - RN^{Pb210} \quad (2.6)$$

The solution to this differential equation (2.2), considering that at the initial moment there is no presence of ^{222}Rn is expressed in the following Eq. (3).

$$N^{Rn222} = [(Q/V_r)/(\lambda^{Rn222} + R)]\{1 - \exp[-(\lambda^{Rn222} + R)t]\} \quad (3)$$

The radioactive activity due ^{222}Rn in disintegrated atoms per second and per unit volume of contaminated air is defined as follows in Eq. (4).

$$Ac^{Rn222} = [(\lambda^{Rn222}Q/V_r)/(\lambda^{Rn222} + R)]\{1 - \exp[-(\lambda^{Rn222} + R)t]\} \quad (4)$$

Considering equations (2.2)–(2.5), in terms of the disappearance of the progeny ($R + \lambda$) it would be possible to add a term R' corresponding to the disappearance by the phenomenon of the plate out, leaving the parentheses of the equations as $(R + \lambda + R')$. Since the radiological activity must decrease by half (Lagarde et al., 2002), the value of the parentheses cited should be doubled, that is, $R' = R + \lambda$.

The mass of radon and its radiological activity ($\text{Bq}\cdot\text{m}^{-3}$) are calculated based on Eq. (3) and Eq. (4) and from the values set out in the Nomenclature. Health administrations from different countries and also the World Health Organization, WHO, set the maximum allowable concentration of radon in air anywhere between 100 and 400 $\text{Bq}\cdot\text{m}^{-3}$. In the case of mechanical ventilation, the amount of the air change rates, R , necessary to ensure that the maximum concentration 400 $\text{Bq}\cdot\text{m}^{-3}$ is obtained from Eq. (5). At the same time, Eq. (5) is obtained from Eq. (4) whereas a time, t , is sufficiently long enough so that the transitional effect disappears and a steady state scenario can be considered. In the present study this steady state condition is reached in six days.

$$400 = (\lambda^{Rn222}Q/V_r)/(\lambda^{Rn222} + R) \quad (5)$$

2.4.2. Case 2: consideration of spatial gradients

ANSYS-CFX is an accepted and widely used finite elements software. With this tool, the geometry of the NORM waste processing plant has been discretized in a mesh of approximately $1.5 \cdot 10^6$ tetrahedrons (Fig. 2). This implies an average size per element of about 5.8 L that guarantees reliable results.

The use of a turbulence or laminar flow model has been considered depending on the Reynolds number, in this study for a number of air change rates greater than 5 the K- ϵ model has been used.

Due to the industrial process developed and when considering the radioactive decays, the temperature of the sources of ^{222}Rn (Scale Silo and Curing Drums) is approximately 333 K. It may seem that a phenomenon of buoyancy that tends to raise the radioisotopes, and is going to take place but it is not like that. This is because the concentration of

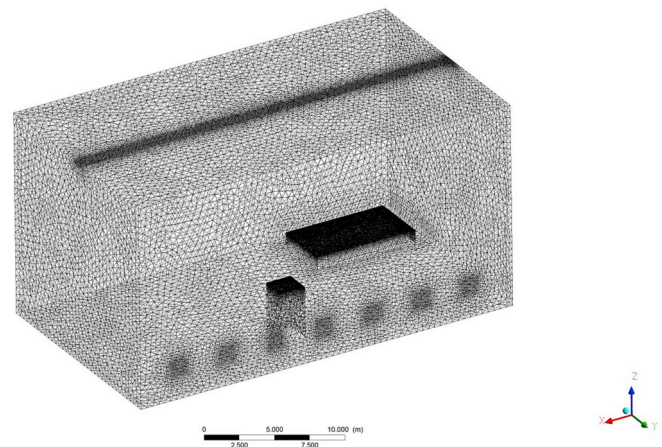


Fig. 2. Mesh diagram.

^{222}Rn gas is so low that this radioactive gas temperature is immediately balanced out to the room temperature. The ^{222}Rn atoms are pulled by the air where buoyancy effects can occur due to differences in temperature caused by air conditioning systems, as is the case in stratifying displacement ventilation systems, not caused by radiological considerations. Thermal loads have not been considered so as not to introduce other variables that affect the conclusions of the work. Energy equation has been neglected, since the process of diffusion of ^{222}Rn and its progeny are considered isothermal, at a 298 K constant temperature.

In the mass conservation equations, ANSYS software allows the introduction of the generation and dissipation terms (source and sink terms, respectively), expressed as chemical reactions of the first order, by means of kinetics constants multiplied by the concentrations of the reactants. Radioactive decay as such is not a chemical reaction; however it can be simulated as such. Thus, a radioisotope like ^{222}Rn dissipates proportionally to its concentration; the proportionality constant is the decay constant, λ .

For each ^{222}Rn dissipated atom, one ^4He atom and one ^{218}Po atoms appear. For the chemical species source and sink terms, ANSYS software requires them to be expressed in terms of mass per unit time per unit volume ($\text{g}\cdot\text{s}^{-1}\cdot\text{m}^{-3}$). Thus, the used expressions for the ^{222}Rn decay are shown in equations (6.1)–(6.3):

$$^{222}\text{Rn} \text{ sink term: } \lambda^{Rn222} [Rn222] M^{Rn222} \quad (6.1)$$

$$^{218}\text{Po} \text{ source term: } \lambda^{Rn222} [Rn222] M^{Po218} \quad (6.2)$$

$$^4\text{He} \text{ source term: } \lambda^{Rn222} [Rn222] M^{He4} \quad (6.3)$$

Where: λ^{Rn222} decay constant of ^{222}Rn ($\ln 2/3.8235$ days = $2.10014 \cdot 10^{-6} \text{ s}^{-1}$), $[Rn222]$ molar concentration of radon ($\text{mol}\cdot\text{m}^{-3}$), M^{Rn222} , M^{Po218} , M^{He4} molecular mass of ^{222}Rn , ^{218}Po and ^4He ($\text{g}\cdot\text{mol}^{-1}$).

Taking the same considerations for the remaining radioisotopes of the simplified chain of disintegration, all the radioisotopes are associated with a source term as follows as shown in equations (7.1)–(7.7):

$$^{222}\text{Rn} \text{ source term: } Q/V_r - \lambda^{Rn222} [Rn222] M^{Rn222} \quad (7.1)$$

$$^{218}\text{Po} \text{ source term: } \lambda^{Rn222} [Rn222] M^{Po218} - \lambda^{Po218} [Po218] M^{Po218} \quad (7.2)$$

$$^{214}\text{Pb} \text{ source term: } \lambda^{Po218} [Po218] M^{Pb214} - \lambda^{Pb214} [Pb214] M^{Pb214} \quad (7.3)$$

$$^{214}\text{Bi} \text{ source term: } \lambda^{Pb214} [Pb214] M^{Bi214} - \lambda^{Bi214} [Bi214] M^{Bi214} \quad (7.4)$$

$$^{214}\text{Po} \text{ source term: } \lambda^{Bi214} [Bi214] M^{Po214} - \lambda^{Po214} [Po214] M^{Po214} \quad (7.5)$$

$$^{210}\text{Pb} \text{ source term: } \lambda^{Po214} [Po214] M^{Pb210} \quad (7.6)$$

$$^4\text{He} \text{ source term: } \lambda^{Rn222} [Rn222] M^{He4} + \lambda^{Po218} [Po218] M^{He4} + \lambda^{Po214} [Po214] M^{He4} \quad (7.7)$$

For the simultaneous resolution of the Fick's diffusion equation, the Navier-Stokes equations and mass conservation equations, the CFD simulation program was used, and the results are shown below. It is assumed that the space is hermetic, and all the supply air enters the seven conduit inlets and the air is extracted via the roof opening, by means of a mechanical ventilation system.

The interaction of the air in the plant with the exhaled radon and its progeny essentially responds to inertial or mechanical forces. In essence, it is a phenomenon of air movement, governed by the Navier-Stokes equations, with one source term from the surfaces of the Curing Drums and Scale Silo, with a condition of input (air inlet in the side-wall) and output (air outlet in the roof), where the atoms of radon and its progeny are pulled through the indoor environment regardless of their nature, like they were gaseous atoms of any other element which comprises air.

Considering the plate out phenomenon, a simple analysis of the problem would be possible by increasing the value of R , so that more

progeny are eliminated via ventilation, thus compensating for the plate out effect. However, this option modifies the real patterns of air velocity, associated turbulence and mixing, introducing error in the calculations. For a real calculation, we must consider the phenomenon of electrostatic adhesion to the walls and dust in suspension, a phenomenon not yet sufficiently defined. As heretofore seen, not including the plate out phenomenon adds a safety factor.

9 different airflow rates have been simulated, namely 0.1, 0.25, 0.50, 0.75, 1, 2.5, 5, 7.5 and 10 air changes per hour, based on the total volume of the NORM waste processing plant.

The code simultaneously solves the Navier-Stokes equations, mass conservation equations and the diffusion equation, after thousands iterations. For the Intel (R) Core (TM) i5-3570 CPU @ 3.40 GHz computer configuration, the computing time is quite variable and depending on the number of air change rates computed. The values oscillate between 52.5 h when $R = 0.1$ and 3.4 h for $R = 10$. A low number of renovations (air change rates) imply lesser turbulence in the space, lower mixing and more pronounced spatial gradients of radioisotope concentration that must be computed iteratively with high computational cost. On the contrary, a higher number of air change rates imply turbulence, mixing, more homogeneous concentrations, and lower gradients of concentration of radioisotopes that are calculated more quickly.

2.5. Computational modelling validation using experimentally measured data

To validate the computational model, experimental measured data of ^{222}Rn concentration in the environment is carried out in a field test enclosure. Subsequently, a CFD imagining of the same field test results is realized, where measurements are compared to the computational analyses so as to verify convergence.

The field test enclosure is a room ($4.10 \times 4.52 \times 3.08$ m) with an air extractor in the upper side of the room and an air intake via an access door in the opposite lower side of the room. An air extract device has been placed, with a flow rate of $6 \text{ m}^3 \text{ h}^{-1}$ which implies approximately 0.1 renovations per hour. Fig. 2 shows the layout and form of the test room, the fan extract position, the air intake, the location of the granite radon emitting dust and the measurement points in the test and simulation.

In the plant case study, there are two sources of radon, Curing Drums and Scale Silo, which together a total emit $1.93 \cdot 10^{12}$ ^{222}Rn atoms $\cdot\text{h}^{-1}$ into the plant. Meanwhile in the test room, there is a single source formed by granite dust, Red-Sayago (Ferrero, 2013). To obtain the radon exhalation rate of granite dust the approach recommended in the IAEA Technical Report 474 (IAEA, 2013) has been used. The amount of this dust dispersed in the test room is such that the ratio of ^{222}Rn atoms exhaled in the test room is the equal to the ratio calculated in the factory simulated plant.

The measuring devices are the Radon Scout SARAD detector. The duration of each measurement was three days, with an integration time of 1 h, totaling approximately 72 concentration values of ^{222}Rn . In addition, the relative humidity and temperature were measured. However in the experimental test, and given the stable environmental conditions of the enclosure (temperature and relative humidity around 286 K and 68%), they are not considered as there are no related variations in concentrations of ^{222}Rn due to temperature-humidity changes. The collected data is downloaded with the software Sarad Radon Vision 6.2.0 and treated with the same program and summarized with an Excel spreadsheet.

As previously discussed, the molar fraction of ^{222}Rn in air is extraordinarily small. This condition results in the hourly measurement values oscillating over a very wide range, therefore it is necessary to consider the average values in a longer timescale. Consequently, in this case study three days were used for averaging values.

Also, the concentration of ^{222}Rn has been measured in the test

facility before introducing the granite dust contaminating source so as to know the background radioactivity concentrations. This measurement was carried out during 237 h and has given the following result: minimum value 0, maximum value 74 Bq·m⁻³, average value 23 Bq·m⁻³ ± 3.9%.

In Table 1, the obtained values of ²²²Rn concentration both in the experimental facility and the simulated program are compared. As shown in the same, the difference between the values obtained from the simulation by CFD and those measured in situ, is of the order of 10%. In view of these results, it is considered that the simulation obtained has a sufficient level of precision to carry out this investigation.

Fig. 3 shows the CFD simulated layout of the test room. At the floor level can be seen the radon emission source, the granite dust of the Red-Sayago rock with the SARAD sensor 1 above the emission source.

3. Results

3.1. Case 1: uniform concentrations

By introducing the values considered in the nomenclature and solving Eq (5), a value of air changes per second, *R*, of 3.28·10⁻⁴ is achieved, i.e. 1.18 air changes per hour in the studied enclosure.

It is noted that this ventilation flow rate used for the perfect dilution ventilation method (uniform spatial concentration), and despite being very low would theoretically be sufficient enough to maintain the controlled radiological activity to a set limit. However, such a low flow rate (air movement) implies very low air velocity and therefore little turbulence and admixing, making it most unlikely that the assumption of perfect dilution. Thus a safety factor is necessary. Increasing *R*, the radiological activity in the space decreases proportionally. Therefore and in practice, the higher the ventilation rate than that calculated for controlling radiological activity the higher safety level achieved.

Comparing the two addends of the exponent, *R* (3.28·10⁻⁴·s⁻¹) and λ_{Rn222} (2.10·10⁻⁶·s⁻¹), it can be observed that the cause of the decrease of radon atoms in the plant is mainly due to ventilation and, to a much lesser extent, radioactive decay, logical circumstance considering that the half-life of ²²²Rn is 3.8235 days and in that time, the atoms of ²²²Rn present have been swept out by the exhaust ventilation extracts.

However and in the case of the radon progenies (radon daughters), and given their very short half-life, their radioactive decay is the principal factor for their disappearance.

This progeny radiological activity can be calculated by integrating the differential equations Eqs. (2.1)–(2.6). Considering the secular equilibrium hypotheses it can be assumed that the half-life of radon is much greater than that of its progeny.

$$(Q/V_r) = (\lambda^{Rn222} + R)N^{Rn222} \tag{8.1}$$

$$\lambda^{Rn222}N^{Rn222} = (\lambda^{Po218} + R)N^{Po218} \tag{8.2}$$

$$\lambda^{Po218}N^{Po218} = (\lambda^{Pb214} + R)N^{Pb214} \tag{8.3}$$

$$\lambda^{Pb214}N^{Pb214} = (\lambda^{Bi214} + R)N^{Bi214} \tag{8.4}$$

$$\lambda^{Bi214}N^{Bi214} = (\lambda^{Po214} + R)N^{Po214} \tag{8.5}$$

From these expressions, the activities of the most significant radioisotopes present in the contaminated air of the factory plant are derived for different air change rates *R* per hour, considering a steady state, see Table 2.

In Table 2, the consistent decrease of the radiological activity for the same radioisotope can be seen as the rate of air change increases. Also, for a same air renewal value, a decrease in activity is observed as the decay chain effect of the progenies continues, further decreasing the longer the half-life. In the case of ²¹⁴Po, its half-life is so small (164.3 μs) that its activity is exactly the same as that of its parent ²¹⁴Bi.

3.2. Case 2: consideration of spatial gradients using computational analysis, CFD

Fig. 4 shows a combined plot of 3 different planes, creating a 3 dimensional imaging of the presence of ²²²Rn and its radiological activity in Bq·m⁻³, in the horizontal plane, Z, at a height of 1.6 m, considered the average height of inhalation in occupied zones, and two vertical planes, while using 0.5 air changes per hour.

Fig. 5 shows a plot of ²¹⁸Po radiological activity (Bq·m⁻³), over the same planes as indicated in Fig. 4 but with 1 air change rate per hour. A high concentration of the apparently isolated ²¹⁸Po radioisotope is observed in the upper zone. This fact is given by the ventilation air velocity pattern, which enters 0.32 m s⁻¹, reaches the drums curing emission source and picks up the radioisotope reaching the wall area, which pushes the polluted air in an upward spiral towards the roof extraction point.

In Fig. 6, the radiological activities of radioisotopes, corresponding to the thick magenta horizontal center line shown in Fig. 5, and where in Fig. 6 the coordinate X = 30.6 m represents the rear facade of the plant, the opposite wall to the door entrance and where X = 0 m represents the main facade where the door access is (see also Fig. 1).

It is also observed that the activity pattern of the different radioisotopes is the same but decreasing as the order of the disintegration chain progresses: the most active radioisotope is ²²²Rn, then ²¹⁸Po, followed by ²¹⁴Pb, and ²¹⁴Pb and then finally ²¹⁴Po. This circumstance, which was already evident in the theoretical model, is corroborated by the simulations and follows the different half-lives of the radioisotopes. Again the case of ²¹⁴Po: its half-life is so small (164.3 μs) that extraction-ventilation does not have time to remove it from the factory plant, so its activity is exactly the same as that of its parent ²¹⁴Bi, overlapping the graphs in Fig. 6. The half-life of ²¹⁸Po is 3.098 min, a small fraction of time compared to 2 h required for the renovation of the plant air, (*R* = 0.5 in the example), so the activity of ²²²Rn and ²¹⁸Po are very similar, somewhat lower ²¹⁸Po in areas of higher concentration of ²²²Rn. The discrepancy seen between the activity of ²²²Rn and ²¹⁸Po around X = 15 m corresponds to the emission of ²²²Rn from the plant source, and there is a clear decrease in the cases of ²¹⁴Pb and ²¹⁴Bi (whose graph is covered by ²¹⁴Po) since their average lives, 26.8 and 19.9 min, are very small time fractions in the 2 h period, reason why the mechanical extraction of these radioisotopes is considerable.

After the post-processing of the 9 simulations, a summary is presented in Table 3 (similar to Table 2). For each air change rate value of *R*, the average activity present in the plant of each radioisotope of the simplified radioactive chain is presented. The values change, according to the number of air change rates, and also depending on the position of the radon source (Curing Drums and Scale Silo), the position of the supply air and its extraction, from the respective areas, and of course considering all the key factors in a ventilation system.

4. Discussion

The effectiveness of decontamination by a ventilation system is clearly reflected in Tables 2 and 3. With lower air change rates, the atmosphere of the plant can be decontaminated. This circumstance is due to the extremely low amount of radon and its progeny present in the plant; hence a relatively low amount of air is able to dilute it.

Table 4 shows the total activity calculated by the perfect dilution

Table 1
Radon radiological activity, experimental method vs. computational.

	Sensors, Measured Radon Activity (Bq·m ⁻³)	Computational Radon Activity (Bq·m ⁻³)
SARAD Sensor 1	285	256
SARAD Sensor 2	97	88

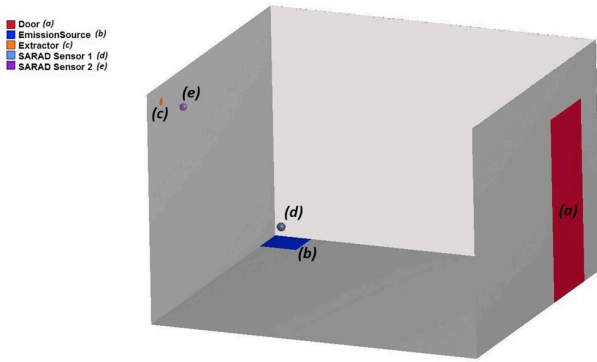


Fig. 3. Test room.

Table 2
Radioactivity of radon and its progeny, perfect dilution.

R (h ⁻¹)	Rn-222	Po-218	Pb-214	Bi-214	Po-214
	Activity (Bq·m ⁻³)	Activity (Bq·m ⁻³)	Activity (Bq·m ⁻³)	Activity (Bq·m ⁻³)	Activity (Bq·m ⁻³)
0	62953	62953	62953	62953	62953
0.1	4421	4389	4123	3935	3935
0.25	1846	1813	1561	1394	1394
0.5	937	903	683	551	551
0.75	628	594	401	295	295
1	472	439	267	181	181
2.5	190	160	61	28	28
5	95	69	16	5	5
7.5	63	41	7	2	2
10	48	27	4	1	1

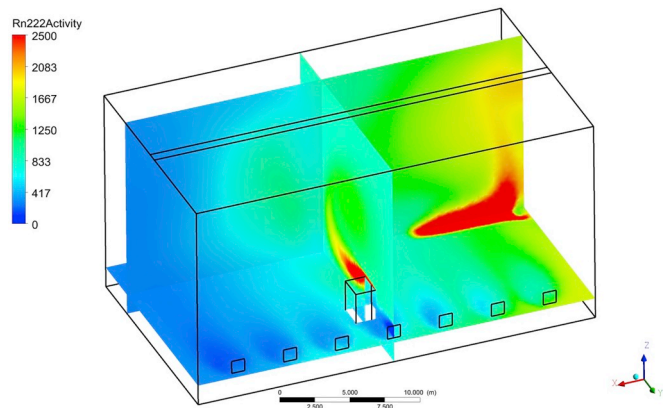


Fig. 4. Horizontal and vertical Plot of ²²²Rn radiological activity; renovations/hour R = 0.5 ACH.

method (from Table 2) -second column (Bq·m⁻³)- and also calculated by CFD method (from Table 3) -third column (Bq·m⁻³)- for different air change rates -first column (h⁻¹)- and an average air velocity -fourth column (m·s⁻¹)-, the average Reynolds numbers -fifth column-are compared.

In Table 4, it is observed that the pollution values calculated by the CFD are greater than those calculated by the perfect dilution method; the difference being ≥ 25%. The concentration values shown for the CFD are calculated by the CFD solver as an average value present in the total volume of the plant and the simulated test room. Also, it is observed that the difference in absolute values between both calculations decreases as the ventilation ratio increases.

This fact is coherent as the air change rate, R, increases, the greater the velocity, turbulence, and intensity of the mixture, bringing the ventilation effect closer to the hypothesis of perfect dilution. As can be

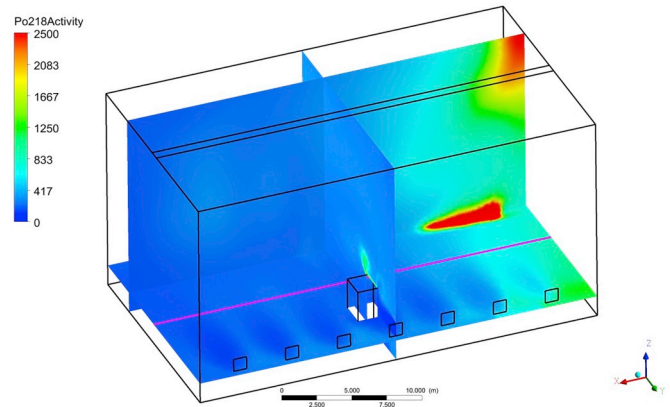


Fig. 5. Horizontal and vertical Plot ²¹⁸Po radiological activity; renovations/hour R = 1 ACH.

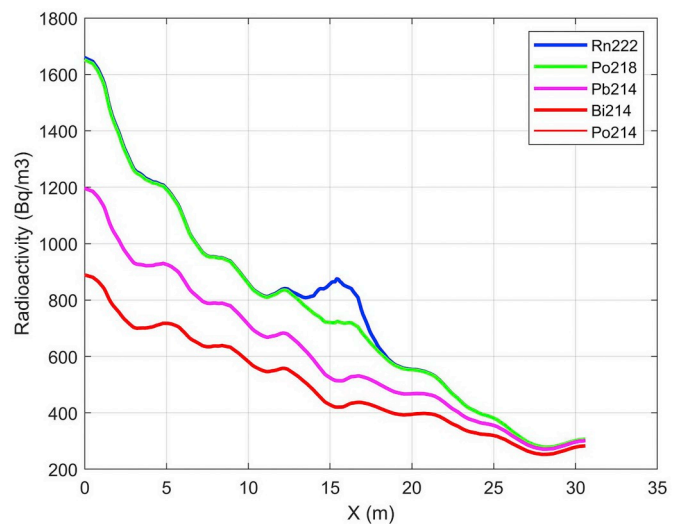


Fig. 6. Radiological activity Chart; renovations/hour R = 0.5 ACH.

Table 3
Average radioactivity of radon and its progeny, spatial gradients of concentration.

R (h ⁻¹)	Rn-222	Po-218	Pb-214	Bi-214	Po-214
	Activity (Bq·m ⁻³)	Activity (Bq·m ⁻³)	Activity (Bq·m ⁻³)	Activity (Bq·m ⁻³)	Activity (Bq·m ⁻³)
0	40520	40520	40520	40520	40520
0.1	5103	5071	4914	4698	4698
0.25	2141	2107	1875	1677	1677
0.5	991	956	743	600	600
0.75	752	677	477	359	359
1	544	508	313	208	208
2.5	200	167	70	33	33
5	110	82	20	6	6
7.5	78	58	9	2	2
10	58	35	5	1	1

seen in Table 4, for turbulent regimes that correspond to Reynolds values higher than 5·10⁵, where the perfect dilution hypothesis is applicable with good results.

However, these approximations are valid for the total averaged mass of radioisotopes present in the air of the plant, not for their spatial distribution, which can present important variations depending on the locations of the radioactivity sources and supply and air extraction points.

Table 4
Comparative of methods, Perfect dilution vs CFD.

R (h ⁻¹)	Total activity (Bq·m ⁻³)	Total activity (Bq·m ⁻³)	Velocity (m·s ⁻¹)	Reynolds number
	calculated by P Dilution method	calculated by CFD method		
0.1	20802	24484	5.2086·10 ⁻³	6.8763·10 ³
0.25	8008	9477	1.4145·10 ⁻²	1.8674·10 ⁴
0.5	3626	3890	5.1633·10 ⁻²	6.8165·10 ⁴
0.75	2213	2624	5.3154·10 ⁻²	7.0173·10 ⁴
1	1540	1781	5.5989·10 ⁻²	7.3916·10 ⁴
2.5	466	503	1.5983·10 ⁻¹	2.1101·10 ⁵
5	190	224	3.4827·10 ⁻¹	4.5978·10 ⁵
7.5	114	149	5.3361·10 ⁻¹	7.0446·10 ⁵
10	80	100	7.1256·10 ⁻¹	9.4072·10 ⁵

For the analysis of a spatial distribution of radioisotopes and also in cases where ventilation systems are low velocity (displacement ventilation systems), the perfect dilution method is not enough in itself as a calculation method to determine radon concentration and therefore the use of a CFD is needed. This is more so the case in displacement ventilation applications, which air characterized by a low turbulent airflow, lower discharge velocities and pronounced thermal gradients due to the buoyancy effect of the heat source.

Although the economic impact of a full CFD analysis could be determining, in this research some guidelines are established so as to reduce costs and simplify calculations, as follows, a simplified method is proposed. The CFD study of radon contamination is essentially a study of fluid mechanics. The mass of radon and its progeny are so extraordinarily low that its presence does not alter the velocity field. Radon and its progeny simply are carried by the air stream as if it was any other gas. Their physical properties such as density or viscosity have no impact in practice. The diffusion coefficients of radon and its progeny in air are only relevant in case of natural ventilation. This study is assumed isotherm. In the case of temperature gradients it is necessary to perform a CFD calculation considering the effect of buoyancy, which requires greater computational investment.

Usually, in industrial applications, the calculation of the absorbed radiation dose is based on the radon concentration and the radon/progeny equilibrium factor. The equilibrium factor of radon and its progeny is influenced by various aspects where the particulate concentration and ventilation rate have a bigger impact. According to the UNSCEAR a typical equilibrium values in dwellings range between 0.2 and 0.8 (UNSCEAR, 2006). The International Commission on Radiological Protection (ICRP, 2009) has agreed an equilibrium factor of 0.4 as a standard international value. However, this CFD method allows the direct calculation of the activities of each and every radioisotope of the progeny, allowing and calculating with greater precision the dose of radiation absorbed.

5. Conclusions

The proposed method in this paper for the analysis of the presence of radon and its progeny can serve as basis for further studies and experimental analysis, for determining the correct ventilation approach and its effectiveness in controlling the presence of radon and its progeny in the breathing zone in occupied spaces, when considering a minimum necessary airflow rate in dilution ventilation applications.

In indoor spaces where NORM waste is handled, radon concentrations and its progeny are harmful to health and above permitted dosage levels by work and safety legislation can be achieved.

The resulting air volumes needed to realize the radiological decontamination of the radon and its progeny are low, and less than those required by the standard ventilation levels for occupational safety and thermal process control. The mass of any other contaminant, of any

other nature, will always be millions or even billions of times the mass of radon in the plant. That is, the necessary ventilation required for radon and its progeny mitigation is less than that required for the thermal treatment and/or the removal of other pollutants.

To reasonably meet the assumption of perfect dilution, frequently used in industrial ventilation applications, it is necessary intensive admixing of the ventilation air and the room air. In case of high air velocities and turbulence the method of perfect dilution can be used with less error. Consequently, it can be assumed that theoretical studies based on the assumption of perfect dilution for radon and its progeny in the air are accurate when the air change per hour, R, is greater than 5.

Given the impossibility of realizing an experimental measurement of radon and its progeny radioactivity on an immediate or short term base, it is very difficult, if not impossible with current technology, to detect the presence of radon. The CFD analysis method is therefore a powerful tool for the design of a ventilation system for a NORM waste processing plant.

Both, the CFD calculation and perfect dilution method, allow for the determining of the activity of each and every one of the radioisotopes of the progeny, which allows a more precise calculation of the absorbed doses by workers in the oil extraction industry, making unnecessary the estimation of the radon/progeny equilibrium factor.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.petrol.2019.106360>.

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