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Radiation Dose Reconstruction of Natural Radioactivity in Fly Ash and Environmental Materials from Morupule a Coal-Fired Power Station in Botswana

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Abstract: ***Dose reconstruction modelling was used in estimating the average annual effective doses due to natural radionuclides to both workers and public members within Morupule A Coal-Fired Power Station and its environs to include the years 1985, 1995, 2005, 2015, 2025, 2035 and 2045. For these years, average fly ash annual effective doses were estimated to be 0.1817 mSv, 0.4591 mSv, 0.7561 mSv, 0.3202 mSv, 0.1826 mSv, 0.1367 mSv and 0.1239 mSv respectively. In the case of coal samples, the values were 0.0695 mSv, 0.1822 mSv, 0.3028 mSv, 0.1258 mSv, 0.0700 mSv, 0.0601 mSv and 0.0462 mSv respectively. For soil samples, they were estimated to be 0.0482 mSv, 0.0907 mSv, 0.1361 mSv, 0.0695 mSv, 0.0484 mSv, 0.0413 mSv and 0.0394 mSv respectively. With regards to water samples, they were estimated to be1.600 µSv, 4.900µSv, 8.300µSv, 3.300µSv, 1.600µSv, 1.100µSv and 1.000 µSv respectively. At the time of sample analysis, the average annual effective doses for the study area were estimated to be 0.320 mSv, 0.126 mSv, 0.069 mSv and 0.003 mSv for fly ash, coal, soil and water samples respectively. All these annual effective doses were due to the natural radionuclides Th-232, U-238 and K-40 in fly ash, coal, soil and water (from the fly ash ponds) samples collected from the study area.All reconstructed average annual effective doses are within the recommended public and occupational dose limits of 1 mSv and 20 mSv respectively. Direct gamma ray spectrometry was used in the initial determination of the above natural radionuclides.***

Keywords: dose reconstruction, gamma spectrometry, fly ash, coal, annual effective dose

1. Introduction

This study has been undertaken to reconstruct the radiation doses from various matrices due to natural radioactivity at Morupule A Coal-Fired Power Station and its environs to include the 60 year period from 1985 to 2045. Morupule A Coal-Fired Power Station is located in the central district of Botswana. An anthropogenic activity such as the combustion of coal for electricity generation in coal-fired power stations has been identified among the likely sources of occupational and public exposure to natural radioactivity. According to USEPA, the fly ash waste generated through coal combustion contains NORM and may release even more natural radioactivity into the environment [USEPA, 2006].A NORM Environmental Impact Assessment was never performed prior to the commissioning of Morupule A Coal-Fired Power Station, implying that the natural radioactivity exposure to the coal-fired power station workers and public members in the vicinity is also unknown.In spite of all these, coal-fired power stations are currently not being regulated for natural radioactivity releases in Botswana. This is of concern since most electricity generation in Botswana is actually done through power stations that burn fossil fuels. Morupule A Coal-Fired Power Station has an output of 4 x 33 MW, uses up to 630, 000 tonnes of bituminous coal each yearand has been in operationsince 1986 with no official baseline or reference data on radioactivity levels.

2. Description of Study Area

The study area is Morupule A Coal-Fired Power Station, located in Morupule (Botswana) at GPS coordinates 22.520˚S27.037˚E, as well as its surroundings within a distance 6 km.

Kgaswe Primary School is located approximately 800 m to the south of the power station at GPS coordinates of 22.530˚S27.038˚E. Palapye village is the nearest village and is located approximately 6 km to the east of Morupule A Coal-Fired Power Station. The land surrounding Morupule A Coal-Fired Power Station is mostly used as a communal grazing area for livestock such as goats, cattle and sheep.

3. Materials and methods

3.1 Sample analysis and dose reconstruction

A total of 8 fly ash samples from the fly ash storage area, 7 bituminous coal samples from the coal storage area, 6 water samples from the fly ash ponds and 9 soil samples were collected from Morupule A Coal-Fired Power Station and its environs. All the samples were analyzed by a High Purity Germanium Detector at the laboratory of the Radiation Protection Institute in Accra, Ghana.

Radioactive decay is a random process, therefore we cannot predict if a single nucleus in a sample will undergo radioactive decay in a given time period. What can be predicted is the average decay behavior for a very large number of similar radionuclides N in a sample. During a small interval of time Δt, ΔN of the atoms undergo radioactive decay [Shultis and Faw, 2007]. The probability for any radionuclide in the sample to decay in time interval Δt is therefore given by $\Delta N/N$.

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The Taylor Series method with numerical derivatives was used to approximate numerical solutions to ordinary differential equations. This Taylor Series method was one of the earliest analytic-numeric algorithms used in the approximation of solutions to ordinary differential equations [Miletics and Moln´arka, 2014]. The exponential term (decay factor) from the radioactive decay law in Equation 12 was first represented by a polynomial of order 4. This is possible since U-238, Th-232 and K-40 have very large half lives being 4.47 x 10^9 years, 1.41 x 10^{10} years and 1.28 x 10^9 years respectively. To approximate the activity concentration of U-238, Bi-214 was used. This was due to the fact that Bi-214 is a daughter product of U-238 and has a relatively short half life of 19.9 minutes compared to that of U-238 [Loureiro, 1987]. In approximating the activity concentration of Th-232, its daughter product Ac-228 was used. Ac-228 has a relatively short half life of 6.1 hours as compared to its parent radionuclide Th-232. The radioactivity build up [Ahmed, 2007; Mayin, 2014] of the daughter product A at any time t is denoted by Equation 1 below:

$$A = A_o(1-e^{-\lambda t}) \quad (1)$$

Where, A_o represents the initial parent radioactivity, λ is the decay constant and t is the decay time. The approximation of the decay factor $e^{-\lambda t}$ to polynomial form is shown below [Mayin, 2014]:

$$P(\lambda t)=P(x) \quad (2)$$

such that

$$P(x)=e^{-\lambda t}=e^{-x} \quad (3)$$

A polynomial is simply a function which can be stated in the form below:

$$P(x) = c_o + c_1x + \ldots\ldots c_nx^n \quad (4)$$

Where, c_o, $c_1\ldots\ldots c_n$ are constant coefficients and n represents the polynomial order for $c_n \neq 0$ over a finite interval [a, b] [Conte and de Boor, 1981]. The Taylor series method of numerical approximations was used to model the non-linear relationship between the average annual effective dose of samples with respect to the elapsed time in years [Smyth, 1998]. Transforming the function e^{-x} to polynomial form made it easier to approximate and minimized the error of approximation. Values of *P(x)* within the range [a, b] were estimated by interpolation while those outside this range were predicted by extrapolation [Smyth, 1998]. Equation 5 below is a representation of the Taylor series [Stroud, 2003]:

$$P(x) = f(x+h)$$
$$= f(x) + hf'(x) + \frac{h^2}{2!}f''(x) + \cdots \frac{h^n}{n!}f^n(x) \quad (5)$$

The exponential function $P(x) = e^{-\lambda t} = e^{-x}$ was approximated about point x = 0 by means of Equation 5 to the polynomial form shown in 4 by means of the following steps:

$$e^{-x}=P(x)=$$
$$f(0)+(x-0)f'(0)+\frac{(x-0)^2}{2}f^2(0)+\frac{(x-0)^3}{6}f^3(0)+\frac{(x-0)^4}{24}f^4(0) \quad (6)$$

such that:

$$f(0) = 1, f'(0) = -1, f^2(0) = 1, f^3(0) = -1, f^4(0) = 1 \quad (7)$$

Substituting the values from (7) into (6) yields the 4th order polynomial approximation of e^{-x} given by Equation 8:

$$e^{-x} = 1 - x + x^2/2 - x^3/6 + x^4/24 = 0.042x^4 - 0.167x^3 + 0.5x^2 - x + 1 \quad (8)$$

MATLAB algorithm was used for the above computations to generate the 4th order Taylor series polynomial of e^{-x}.

The half life of Bi-214 was used to calculate $x = \lambda t$ for U-238, while the half life ofAc-228 was used in the case of Th-232. The results were then evaluated by utilizing the polynomial approximation expression in (8) above.The activity concentrations of these radionuclides were then reconstructed by means of the radionuclide decay expression below:

$$A = A_o e^{-\lambda t} \quad (9)$$

The annual effective dose due to the samples was calculated. The approximation from Equation 8 was then used in interpolating the growth for the calculated activity concentration of all the samples utilizing the half lives of Bi-214 and Ac-228, being the daughter radionuclides of U-238 and Th-232 respectively. The growth was estimated to include the period of 30 years before and 30 years after the time of sample analysis.

Results and discussion

Reconstructed doses from the study area

Annual effective doses were reconstructed for all samples from the study area. Tables 1 to 4 shows the reconstructed annual effective doses due to fly ash, coal, soil and water (from the fly ash ponds) samples respectively. Figures 2, 3, 4 and 5 show the dose reconstruction model graphs that represent the results from Tables 1, 2, 3 and 4 respectively. The mean annual effective doses from these tables were used as inputs for these dose reconstruction models.

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Table 1: Reconstructed annual effective doses for fly ash samples

RECONSTRUCTED ANNUAL EFFECTIVE DOSES FOR FLY ASH STORAGE AREA (mSv/y)

	1985	1995	2005	2015	2025	2035	2045
ASH-1	0.1947	0.4946	0.8156	0.3445	0.1958	0.1462	0.1323
ASH-2	0.1993	0.5030	0.8279	0.3510	0.2005	0.1502	0.1361
ASH-3	0.1926	0.4869	0.8019	0.3397	0.1937	0.1450	0.1314
ASH-4	0.1772	0.4488	0.7395	0.3129	0.1783	0.1333	0.1208
ASH-5	0.1856	0.4673	0.7687	0.3263	0.1867	0.1401	0.1270
ASH-6	0.1827	0.4592	0.7551	0.3208	0.1837	0.1380	0.1252
ASH-7	0.1497	0.3860	0.6388	0.2678	0.1507	0.1116	0.1007
ASH-8	0.1707	0.4268	0.7008	0.2986	0.1716	0.1292	0.1174
Min.	0.1497	0.3860	0.6388	0.2678	0.1507	0.1116	0.1007
Max.	0.1993	0.5030	0.8279	0.3510	0.2005	0.1502	0.1361
Mean	0.1817	0.4591	0.7561	0.3202	0.1826	0.1367	0.1239
Std Dev.	0.0159	0.0388	0.0633	0.0273	0.0160	0.0122	0.0112

Table 2: Reconstructed annual effective doses for coal samples

RECONSTRUCTED ANNUAL EFFECTIVE DOSES FOR COAL STORAGE AREA (mSv/y)

	1985	1995	2005	2015	2025	2035	2045
Coal 1	0.0585	0.1535	0.2550	0.1060	0.0589	0.0432	0.0389
Coal 2	0.0623	0.1610	0.2667	0.1116	0.0627	0.0464	0.0418
Coal 3	0.1004	0.2681	0.4476	0.1843	0.1012	0.1344	0.0657
Coal 4	0.0959	0.2558	0.4269	0.1759	0.0967	0.0702	0.0629
Coal 5	0.0565	0.1457	0.2410	0.1010	0.0569	0.0422	0.0381
Coal 6	0.0592	0.1516	0.2505	0.1054	0.0596	0.0443	0.0400
Coal 7	0.0536	0.1398	0.2320	0.0967	0.0540	0.0398	0.0358
Min.	0.0536	0.1398	0.2320	0.0967	0.0540	0.0398	0.0358
Max.	0.1004	0.2681	0.4476	0.1843	0.1012	0.1344	0.0657
Mean	0.0695	0.1822	0.3028	0.1258	0.0700	0.0601	0.0462
Std Dev.	0.0198	0.0550	0.0927	0.0374	0.0200	0.0343	0.0125

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Table 3: Reconstructed annual effective doses for soil samples

RECONSTRUCTED ANNUAL EFFECTIVE DOSES FOR SOIL IN AND AROUND MORUPULE A COAL-FIRED POWER STATION (mSv/y)

	1985	1995	2005	2015	2025	2035	2045
Soil 1	0.0424	0.0793	0.1188	0.0609	0.0426	0.0365	0.0348
Soil 2	0.0474	0.0874	0.1302	0.0674	0.0475	0.0409	0.0391
Soil 3	0.0413	0.0767	0.1145	0.0590	0.0415	0.0357	0.0340
Soil 4	0.0466	0.1031	0.1634	0.0748	0.0469	0.0375	0.0349
Soil 5	0.0431	0.0845	0.1288	0.0638	0.0433	0.0365	0.0346
Soil 6	0.0383	0.0632	0.0899	0.0507	0.0384	0.0342	0.0331
Soil 7	0.0581	0.1158	0.1777	0.0870	0.0583	0.0488	0.0461
Soil 8	0.0681	0.1161	0.1674	0.0921	0.0683	0.0604	0.0582
Soil 9	0.0486	0.0902	0.1346	0.0694	0.0488	0.0420	0.0400
Min.	0.0383	0.0632	0.0899	0.0507	0.0384	0.0342	0.0331
Max.	0.0681	0.1161	0.1777	0.0921	0.0683	0.0604	0.0582
Mean	0.0482	0.0907	0.1361	0.0695	0.0484	0.0413	0.0394
Std Dev.	0.0094	0.0179	0.028	0.0133	0.0094	0.0084	0.0082

Table 4: Reconstructed annual effective doses for water samples

RECONSTRUCTED ANNUAL EFFECTIVE DOSES FOR WATER SAMPLES FROM THE ASH PONDS (mSv/y)

	1985	1995	2005	2015	2025	2035	2045
Water 1	0.0011	0.0032	0.0055	0.0022	0.0011	0.0008	0.0007
Water 2	0.0011	0.0031	0.0052	0.0021	0.0011	0.0008	0.0007
Water 3	0.0013	0.0040	0.0069	0.0027	0.0013	0.0009	0.0008
Water 4	0.0035	0.0086	0.0140	0.0060	0.0035	0.0027	0.0024
Water 5	0.0007	0.0020	0.0033	0.0014	0.0007	0.0005	0.0005
Water 6	0.0020	0.0083	0.0149	0.0052	0.0021	0.0011	0.0008
Min.	0.0007	0.0020	0.0033	0.0014	0.0007	0.0005	0.0005
Max.	0.0035	0.0086	0.0149	0.0060	0.0035	0.0027	0.0024
Mean	0.0016	0.0049	0.0083	0.0033	0.0016	0.0011	0.0010
Std Dev.	0.0010	0.0028	0.0049	0.0019	0.0010	0.0008	0.0007

Annual effective dose model of the fly ash storage area

The sixty (60) year interpolative and extrapolative annual effective dose model for the fly ash storage area is presented in Figure 2 and represented by the 4th order polynomial:

$$Y = -0.032z^4 + 0.23z^3 - 0.076z^2 - 0.46z + 0.42$$

The model is standardized by the z-score given below, which determines the number of standard deviations the x-axis value (time in years) is from the mean[Larsen and Marx, 2000]:

$$z = (x - 2000)/ 22$$

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Where, z is the z-score, 2000 is the average year, 22 is the standard deviation and x is the predictor data or year of interest. The model utilizes the least squares method which connects data points by means of a best fit line [Hastie, Tibshirani and Friedman, 2009].This model is a reasonable predictor the annual effective dose for the time range $1985 \leq x \leq 2045$.This model predicts alow mean annual effective dose of 0.1817 mSv/year which serves as the average baseline (reference) annual background radiation for 1985/86. This may be attributed to the fact that Morupule A Coal-Fired Power Station began operating in 1986 [UNSCEAR, 2008].

The average annual effective dose then gradually rose to a maximum value of 0.7561 mSv/y in 2005. This may be attributed to an increase in the activity of the coal-fired power station which led to an increased fly ash production [Organo and Fenton, 2008]. The model further predicts a decrease in the average annual effective dose to a value of 0.1239 mSv/year in 2045. This decrease may be attributed to exponential decay according to the Radioactive Decay Law [Benedict, 2012]. The mean annual effective doses estimated by the model are much lower than the public annual effective dose limit of 1mSv [IAEA, 2003].

Annual effective dose model of the coal storage area

The eighty (80) year interpolative and extrapolative annual effective dose model for the coal storage area is presented in Figure 3 and represented by the 4th order polynomial:

$$Y = -0.018z^4 + 0.093z^3 - 0.02z^2 - 0.18z + 0.16$$

The model is standardized by the z-score given below, which determines the number of standard deviations the x-axis value (time in years) is from the mean [Larsen and Marx, 2000]:

$$z = (x - 2000)/ 22$$

Where, z is the z-score, 2000 is the average year, 22 is the standard deviation and x is the predictor data or year of interest. The model utilizes the least squares method which connects data points by means of a best fit line [Hastie, Tibshirani and Friedman, 2009]. This model is a reasonable predictor the annual effective dose for the time range $1985 \leq x \leq 2065$. This model predicts a low mean annual effective dose of 0.0695 mSv/year which serves as the average baseline (reference) annual background radiation for 1985/86. This may be attributed to the fact that Morupule A Coal-Fired Power Station began operating in 1986 [UNSCEAR, 2008].

The average annual effective dose then gradually rose to a maximum value of 0.3028 mSv/y in 2005. This may be attributed to an increase in the activity of the coal-fired power station which led to an increased accumulation of raw coal fuel in the coal storage area[Organo and Fenton, 2008]. The model further predicts a decrease in the average annual effective dose to a value of 0.0269 mSv/year in 2069. This decrease may be attributed to exponential decay according to the Radioactive Decay Law [Benedict, 2012]. The mean annual effective doses estimated by the model are much lower than the public annual effective dose limit of 1mSv [IAEA, 2003].

Annual effective dose model for soil samples from the study area

The sixty (60) year interpolative and extrapolative annual effective dose model forsoil from the study area is presented in Figure 4 and represented by the 4th order polynomial:

$$Y = -0.005z^4 + 0.036z^3 - 0.012z^2 - 0.07z + 0.084$$

The model is standardized by the z-score given below, which determines the number of standard deviations the x-axis value (time in years) is from the mean [Larsen and Marx, 2000]:

$$z = (x - 2000)/ 22$$

Where, z is the z-score, 2000 is the average year, 22 is the standard deviation and x is the predictor data or year of interest. The model utilizes the least squares method which connects data points by means of a best fit line [Hastie, Tibshirani and Friedman, 2009]. This model is a reasonable predictor the annual effective dose for the time range $1985 \leq x \leq 2045$. This model predicts a low mean annual effective dose of 0.0482 mSv/year which serves as the average baseline (reference) annual background radiation for 1985/86. This may be attributed to the fact that Morupule A Coal-Fired Power Station began operating in 1986 [UNSCEAR, 2008].

The average annual effective dose then gradually rose to a maximum value of 0.1361 mSv/y in 2005. This may be due to an increased amount radionuclides in the chimney gases reaching ground by either wet or dry deposition[Szefer and Nriagu, 2006]. The model further predicts a decrease in the average annual effective dose to a value of 0.0394 mSv/year in 2045. This decrease may be attributed to exponential decay according to the Radioactive Decay Law [Benedict, 2012]. The mean annual effective doses estimated by the model are much lower than the public annual effective dose limit of 1mSv [IAEA, 2003].

Annual effective dose model for water samples from the fly ash ponds

The sixty five (65) year interpolative and extrapolative annual effective dose model for water from the ash ponds is presented in Figure 5 and represented by the 4th order polynomial:

$$Y = -0.00037z^4 + 0.0028z^3 - 0.0009z^2 - 0.0054z + 0.0044$$

The model is standardized by the z-score given below, which determines the number of standard deviations the x-axis value (time in years) is from the mean [Larsen and Marx, 2000]:

$$z = (x - 2000)/ 22$$

Where, z is the z-score, 2000 is the average year, 22 is the standard deviation and x is the predictor data or year of interest. The model utilizes the least squares method which

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connects data points by means of a best fit line [Hastie, Tibshirani and Friedman, 2009]. This model is a reasonable predictor the annual effective dose for the time range 1985 ≤ x ≤ 2050. This model predicts a low mean annual effective dose of 0.0016 mSv/year which serves as the average baseline (reference) annual background radiation for 1985/86. This may be attributed to the fact that Morupule A Coal-Fired Power Station began operating in 1986 [UNSCEAR, 2008].

The average annual effective dose then gradually rose to a maximum value of 0.0083 mSv/y in 2005. This may be due to an increased amount radionuclides in the chimney gases reaching ground and ash ponds by either wet or dry deposition [Szefer and Nriagu, 2006]. It could also be attributed to more of the fly ash produced being added to water in the ash ponds [Skodras et al., 2007]. The model further predicts a decrease in the average annual effective dose to a value of 0.0013 mSv/year in 2050. This decrease may be attributed to exponential decay according to the Radioactive Decay Law [Benedict, 2012]. The mean annual effective doses estimated by the model are much lower than the public annual effective dose limit of 1mSv [IAEA, 2003].

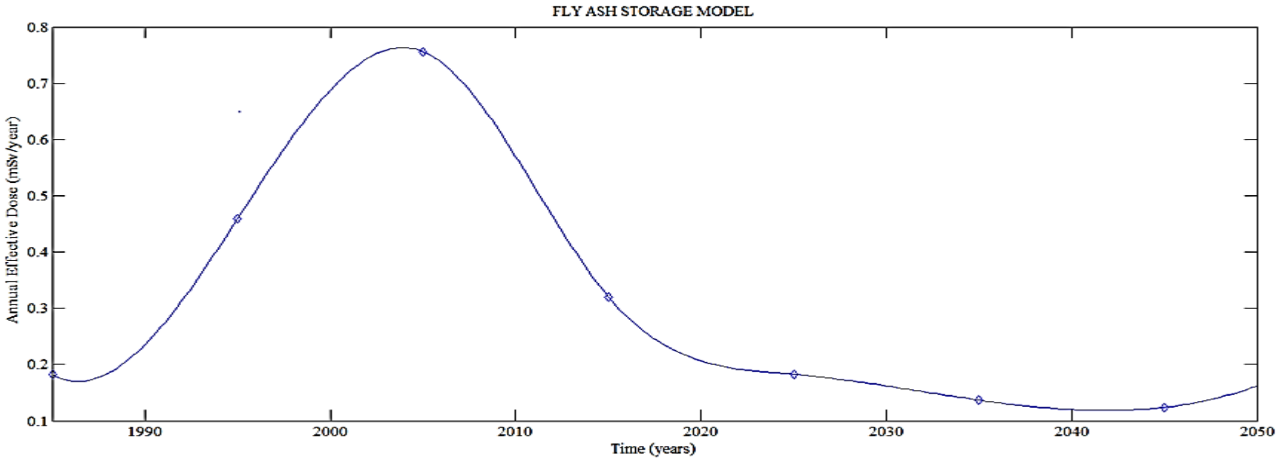

Figure 2: Grapical representation of the fly ash storage area model

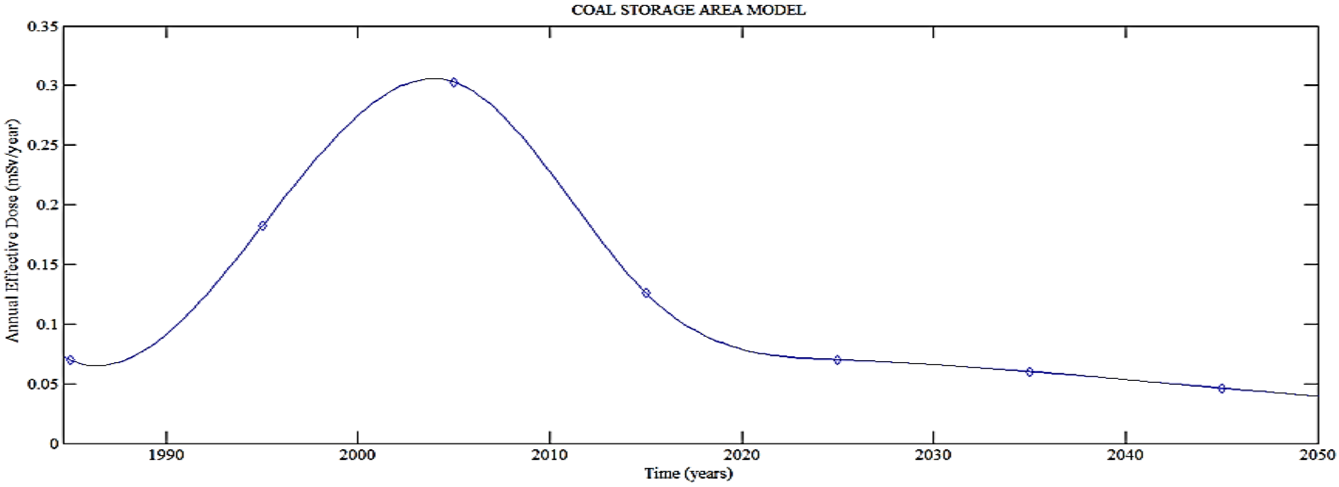

Figure 3: Grapical representation of the coal storage area model

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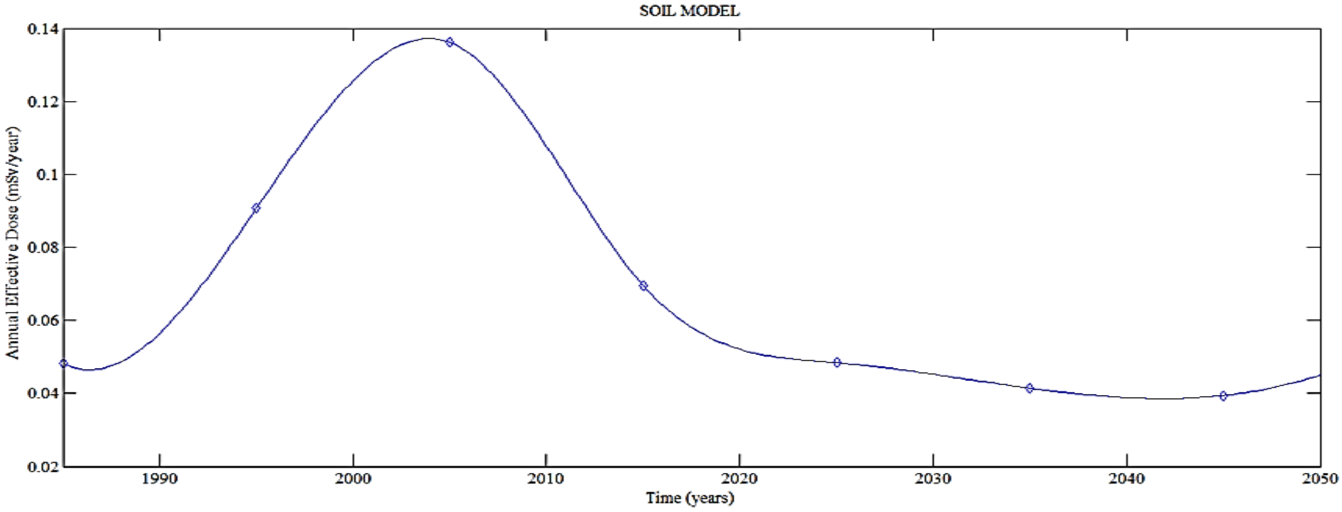

Figure 4: Grapical representation of the soil model for the study area

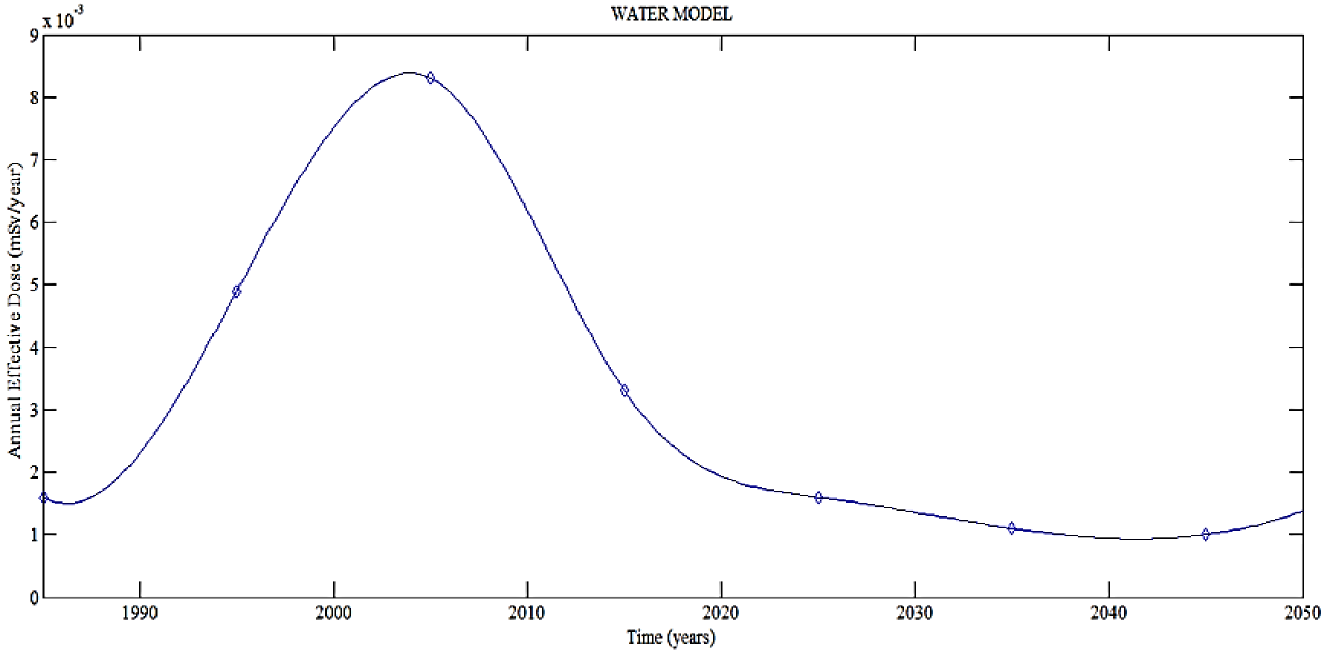

Figure 5: Grapical representation of the water model for the fly ash ponds

4. Conclusions

The average annual effective doses from the study area due to natural radioactivity were estimated to be 0.320 mSv/year, 0.126 mSv/year, 0.069 mSv/year and 0.003 mSv/year for the fly ash, coal, soil and water samples respectively. Through this work, baseline data for the natural radionuclides U-238, Th-232 and K-40 has been estimated by means of a graphical mathematical dose reconstruction modelling to all the study samples. The dose reconstruction model from this work was used to reconstruct radiation doses due to these natural radionuclides in the samples to include the period from 1985 to 2045. Across all samples, the model predicted a very low annual effective dose in 1985/86 and this corresponds to the time when Morupule A Coal-Fired Power Station started operating. The model shows that the annual effective dose gradually increased to a maximum value in 2005 and then eventually decayed off to lower values for all samples. The model used utilized the least squares method which connects data points by means of a best fit line [Hastie, Tibshirani and Friedman, 2009]. For all samples, the estimated and reconstructed mean annual effective doses are much lower than the recommended annual effective dose limit for members of the public and that for occupationally exposed workers, whose values are 1 mSv/year and 20 mSv/year respectively [IAEA, 2003].

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