# Article

**Open Access** 

DELINEATION OF CARBONATE RADIOACTIVE ELEMENTS FROM SOUTHEASTERN NETHERLANDS USING GAMMA- RAY SPECTROMETER

Oghenero Ohwoghere-Asuma

Vrije Universiteit (VU) Amsterdam, Faculty of Earth and Life Science, De Boelelaan 1085, 1081 HV Amsterdam, The Netherlands

Received June 29, 2017; Accepted October 10, 2017

#### Abstract

A Gamma-ray spectrometer (Brand GR410 with GP-21 detector of Geomatrix Exploranium Company) was used to determine the concentrations of Uranium (U), Potassium (K) and Thorium (Th) in limestone deposit exposed at the Curfs quarry in Limburg, Southeastern Netherlands. The concentrations of K in part per million (ppm) ranged from 63 to 102, U from 43-84 while Th from 4 to 13 respectively. The concentration in ppm of Th appeared lowest, followed by U and lastly by K which has the highest values among the three elements. The probable source of Th, and K have originated from clay minerals in the limestone. The origin of U though not clear but it was adduced to the presence of U-rich detrital content of clays. Correlation of the three sections logged showed slight variation in the concentration of Th, K and U across the quarry. The variations are due to the effects of non-homogenous mixing of carbonate sediments by bioturbation.

*Keywords:* Gamma – ray; uranium; potassium; thorium; radioactive.

## 1. Introduction

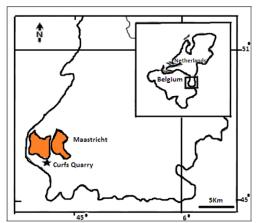
Gamma-ray log has been an essential tool in the exploration and exploitation of hydrocarbon involving siliciclastics and carbonate rocks respectively. It has been and continued to be use in the delineation of lithology, especially in the oil industry. It has found specific application in the discrimination of reservoir rocks from non-reservoir rocks, sandstone from clay and limestone from clay. Environment of deposition of rocks are often deduce from signature of gamma- ray log. The success is attributable to the available of radioactive elements in these rocks. Major radioactive elements that are utilize for well logging are potassium (k), thorium (Th), and Uranium (U). The radioactivity of these elements are often measured in part per million (ppm) by Scintillometry in well logging. Deduction of lithology is by correlating plotted gamma-ray values of K, Th and U in ppm against depth. These radioactive elements maybe interpreted on the bases of their percentages in clays and U- rich detrital minerals contents of clays and U- rich organic matters.

The studied of the radioactivity of organic rich palegic sediments from the Aptian - Albian in the Fucoid Marl Formation in the Marche-Ombrie Basin (Italy) by <sup>[1]</sup> revealed that both the concentration of K and Th signified the presence of clays in limestone. They found it difficult to correlate the presence of U with the abundance of organic matters in black shale alone but also the thickness of the black shale.

The present study is aimed at determine the spatial distribution of radioactive K, Th and U in shallow water carbonate platform exposed at the Curfs quarry in Limburg Southeastem Netherlands, as no previous study exist.

## 2. The study area

The quarry is situated in Southeastern area of Limburg with latitude  $50^{\circ} 45^{1}17^{11}$  N and longitude  $60^{\circ}15^{11}$ E as coordinates. In the south and west bounded by Belgium, in the east by



Germany, partly in the west by Noord-Brabant and Geldland to the north (Figure 1). The area is unique when compared to other regions in the Netherlands in having a carbonate deposit. The presence of carbonate deposit in Maastricht has in essence revealed that Netherlands was formally subtropical to tropical in the time past as depicted by the fossil assemblage of the limestone

Fig.1. Map of the Limburg area, showing the study area <sup>[2]</sup>

## 3. Geologic setting

The Late Kimmirian rifting characterized by proliferation of heat flow, magmatic episodes and faulting led to evolution of several basins in the Netherlands. During the Late Cretaceous, the rifted basin experienced cooling and sea level rose. Coeval episodes of Austrian tectonic led to abrupt culmination of sea level rise in the early Albian. Thereafter, the sea started to rise again in the later part of Albian, the effect affected the entire region of Northwestern Europe <sup>[3]</sup>. At the time of Late Cretaceous, input of basin fills diminished due to source area flooding. This flooding initiated the growth of the limestone deposit. During Paleocene, the basin fills experienced asymmetrical subsidence and the Laramide inversion (Table 1).

At this time, normal faults became reverse faults, erosion of exhumed basin fills and culmination of carbonate sedimentation were some of the effects of the inversion. The cause of the inversion controversially adduced to the Alpine compression and the extent of impact was unknown. A regional extent suggested by <sup>[4-5]</sup>, local inversion <sup>[6]</sup> and <sup>[7]</sup> said there was no convincible evidenced of inversion. The carbonate deposit finally evolved following sea level fall precipitated by regional deformation of the lithosphere <sup>[8]</sup>. The various tectonic phases summarized and presented in Table 1.

Geologically, two distinguishable lithofacies characterized the carbonate outcrop in the Limburg. These are hard and soft. The hard carbonate assumed its hardness by undergoing strong lithification and erosion after deposition, this facies is hardground. It is quite rich in fossil assemblages including sponges, bivalves, bryozoans, brachiopod and serpulids. It also lacks sedimentary structures caused by bioturbation. The grains of the hardgrounds are fining upward, are poorly sorted and moderately cemented. They are conspicuously many and tend to alternate each other. The other facie sandwiched between the hardgrounds is friable coarse-grained sediments with no obvious cementation. Within the layer are depression fills, which are coarse and similar to storm-like- deposit. This storm-like deposit terminates against hardgrounds, which resembles sedimentary pitch out. The fossil assemblage is similar to those of the hardgrounds but with slight variation in the presence of echinoderms, corals and benthic foraminifera. In terms of their velocity property, these two layers are referred to as low (soft) and high (hardground) velocity layers <sup>[9]</sup>.

The colour of the carbonate rock in the quarry varied from slightly yellowish through green to brown. These colours are due to the presence of glauconite and iron minerals in the rock. Sedimentary structures are conspicuously lacking due to the bioturbation, however, synsedimentary structure cross bedding, karst, bioturbation and burrows are common. The variation in the sizes of fossils suggested that low to moderate energy was responsible for deposition <sup>[10]</sup>. Subtropical/tropical and shallow environment of deposition inferred from the assemblages of fossils. Table 1. Tectonic phases and the main event from Late Jurassic to earliest Tertiary in the Netherlands according to [5]

Chronostratigraphy		Group	Tectonic phase	Events	
Early Tertiary	Paleocene	Lower North Sea	Laramide	Renewal of inversion, local erosion, minor rifting and wide spread carbonate sedimentation	
	Maastrichtian Campanian				
Late Cretaceous	Santonian	Chalk	Subhercyian	Strong inversion and erosion of upper Jurassic depoceters; regional differential subsidence. Onset of widespread carbonate sedimentation	
	Conacian				
	Turonian Cenomanian				
	Albian Aptian				
	Barremian		Austrian	Unconformity; local uplift, followed by	
Early Cetaceous	Hauterivian	Rijnland	Late Kimmerian	Erosion and regional Albian transgression	
	Valanginian		Phase II	Onset of Cretaceous transgression over	
	Ryazanian			Late Kimmerian unconformity Diminishing rifting activity,	
				thermal Subsidence	
Late Jurassic		Schieland/	Late Kimmerian	Major rifting and different subsidence, rapid sedimentation	
		Nedersachsen		over	
		(Scruff)	Phase I	Mid Kimmerian Unconformity in	
				narrow and restricted basin	
				local volcanism	

## 4. Material and method

The limestone deposit belongs to the Geulhem Member of the Houthem Formation exposed at the Curfs quarry in the province of Limburg, southeastern, Netherlands. The gamma ray spectrometer cleaned to remove impurity from the exposed surface. The gamma ray spectrometer (Brand GR410 with GP-21 detector of (Geomatrix Exploranium Company) was strongly and firmly held against the outcrop wall to measured natural radiation of carbonate at a spacing interval of 10cm and a total depth of 100cm was covered. Measurement was done and count rate were recorded at an interval of two minutes and the concentration of the potassium, thorium and uranium were recorded in part per million (ppm). Subsequently, the individual concentrations in ppm and the total concentration plotted against depth to obtain the signature pattern of the logged sections. Three sections logged together with spacing distance of 100m apart in-between section. In order to ascertain spatial distribution of radioactive elements across the quarry, sections correlation carried out.

## 5. Results and discussion

Table 2 depicts all sections logged with the gamma-ray spectrometer showed that the concentrations of P, Th and U across the quarry decrease and increase in an alternate manner

from the bottom to the top with more of decrease trend (Figure.2). The concentration of K is relatively higher in the first 40cm and decreases in a trend that is not serially. The concentrations of K at every depth are higher than, those of Th which appeared lower. The concentration of U and K appeared almost same both vertically and laterally across sections logged with slight difference in concentrations, but K concentrations were higher than U.

Depth	Radioactive elements measured					
	Total content	K (ppm)	U (ppm)	Th (ppm)		
100	6.1	0	3.3	0.8		
	784	67	69	10		
90	5.6	0.1	2	0.5		
	723	63	43	6		
80	6.1	0.1	2.4	0.6		
	776	69	51	7		
70	6.1	0	3	0.6		
	777	65	63	8		
60	6.3	0.1	2.3	1.1		
	809	75	51	11		
50	6.6	0	2.7	1.2		
	843	72	59	13		
40	7.1	0	4.1	0.3		
	910	97	84	6		
30	7.1	0.1	4	0.6		
	910	102	84	9		
20	7	0.1	2.7	0.7		
	899	86	57	9		
10	7	0.1	3.5	0.1		
	890	97	72	4		

Table 2. Concentration of radioactive elements in part per million (ppm) obtained from the Curfs quarry in Limburg, Southeastern Netherlands

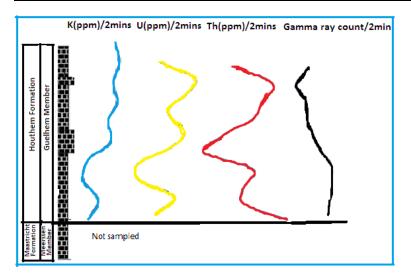


Figure.2. Variation of K, U and Th contents measured by Gamma ray spectrometer counts

The spatial distribution of radioactive elements ascertained by correlating Gamma ray spectrometer values obtained at the different sections logged across the Curfs quarry with one another. The correlation reveals that spatial and temporal distribution of potassium, thorium and uranium reflects almost uniform pattern across the quarry (Figure. 3).

The lateral continuity of hardground in the Curfs quarry correlates with the concentration of radioactive elements across the three sections logged. The only difference is that sections one and two correlated to be at same depth than that section three, which appeared slightly higher. That is, they almost have the same amount of concentration of radioactive elements at different depths. There is no significant distinction between the radioactive elements to the lithology since the limestone deposit is homogeneous.

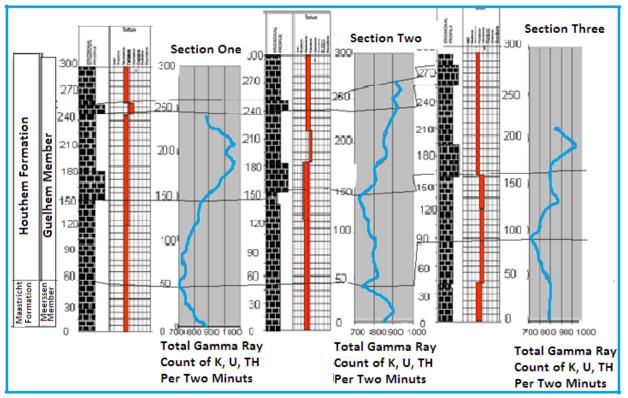


Figure. 3. Correlation of the spatial distribution of K, U, and Th across the Curfs quarry in Limburg, Southeastern Netherlands.

The source of these radioactive elements is determine from the work of others including the work of <sup>[11]</sup>; they studied carbonate rock of the Marche-Ombrie Basin in Italy and <sup>[11]</sup>] studied the carbonate of Gorges du Nan of the Vercors Massive (Western Alps). In the different studies, they claimed that the clays are the precursor of radioactive elements often detected in gamma ray logging equipment such as the one used in the study. More also, an earlier sedimentological study of same Limburg limestone deposit by <sup>[12]</sup> revealed the presence of K rich smectite and high content of authigenic smectite as the origin of clay minerals. These studies supported that the presence of radioactive K and Th in limestone is probably due to availability of clay minerals in limestone. However, evidence available these works, the present study agreed with the result revealed in previous studies. The source of Thorium (Th) and Potassium (K) concentrations in the limestone deposit of Limburg, southeastern Netherland is probably the presence of clays minerals contained in the limestone but needs further clarifications in the future.

The source of uranium in limestone remained an issue yet to be resolved because its' origin, which is often tie to both organic matter and U- rich detrital minerals content of clays in limestone. Thus, the utilization of gamma ray logging equipment in this study may not be sufficient to attribute source of these radioactive elements to clays. Further study is therefore required to ascertain the origin of uranium in the Limburg limestone. However, considering the presence of clays in the study area, the source of Uranium in the Curfs quarry limestone deposit maybe connected to leaching of Uranium rich meteoric water from clay. The presence

of meteoric cements, dissolution of bioclasts and karsts in the quarry observed from petrographic and field observations (another study) is in conformity with the above assertion. Uranium may also been contemporaneously brought in along with limestone sediments. Petrographic analysis revealed the presence of detrital glauconite in peloidal grains, bioclasts and in the pore spaces between bioclasts supports this. The spatial variation in the concentration in the amount of K, Th, and U may have resulted from the bioturbation mixing and reworking of sediments that characterized the Curfs quarry. Non- uniform bioturbation may have contributed to the slight variation in the concentration of the radioactive elements. However, the concentrations of K, Th, and U obtained are relatively lower when compared to most detrital rocks <sup>[10]</sup>, an indication of abundance of clay minerals in siliciclastics than carbonates.

## 6. Conclusions

The presence of radioactive elements of Potassium, Thorium and Uranium in part per million (ppm) was determined in the field with the aid of gamma ray spectrometer and the distribution of these elements both vertically and laterally in the Curfs quarry has no relationship with lithology as the Curfs quarry is made up of homogenous limestone except the clay overlying it that was not shown in the outcrop.

Correlation still remains the only viable means to ascertain the spatial distribution of radioactive elements of potassium (K), Thorium (Th) and uranium in both silliciclastics and carbonates rocks. The variation of these elements observed in the study may have been due to non-uniform mixing of sediments by bioturbation, which characterized the Curf quarry limestone deposit.

The source of radioactive elements of potassium, thorium and uranium are probably link to the presence of clays minerals in limestone. The source of uranium in carbonate rocks is still debatable because it can either be associated with the presence of clays mineral or organic matter present in limestone. The dual nature of having clays mineral and organic matter as source of uranium needs further investigation.

## Acknowledgement

Many thanks to Delta State university Abraka for granting the leave, and the academic and Laboratory staffers of the Tectonic and Sedimentology department of the Faculty of Life and Earth Sciences, Vu University Amsterdam, The Netherlands for the opportunity that enabled the study.

## References

- Fiet N Gorin GE. Gamma ray spectrometer as a tool for stratigraphic correlation in the carbonate-dominated, organic rich pelagic Albian sediments in central Italy, Ecolog. Geo Helv. 2000; 93: 175-181.
- [2] Schioler, P. Brinkhuis H Roncaglia L. Wilson GJ. Dinoflagellate biostratigraphy and sequence stratigraphy of the type Maastrichtian (upper Cretaceous, ENCI, The Netherlands. Marine Micropaleontonlogy, 1997; 31: 65-95.
- [3] Crittenden S. The Albian transgression in the southern North Sea basin, J. Pet. Geol., 1987; 19: 395-414.
- [4] Ziegler PA. Geological Atlas of Western and Central Europe. Shell International Petroleum Maatschappij BV, distributed by Elsevier Amsterdam, 1982. Pp.130.
- [5] Ziegler PA. Geological Atlas of Western and Central Europe (2<sup>nd</sup> edition) Shell International Petroleum Maatschappij BV, Geological Society Publishing house (bath); 1990; pp.230.
- [6] Baldschuhn, R., Best, Kockel, F inversion tectonic in the northwest German basin; in spencer, A M (ed); generation and accumulation and production of Europe hydrocarbon. Special publication, European association of petroleum geologist oxford university press 1991; 1: 145-159.
- [7] Kockel, F. Inversion structures in central Europe expression and reasons; an open discussion, Netherlands J. Geosci. / Geologie en Mijnbouw, 2003. 82: 351-366.
- [8] Coetingh, S Intraplate stresses; a new tectonic mechanism for fluctuation of relative sea level. Geology, 1986; 14:617-620.

- [9] Ohwoghere-Asuma O. Acoustic velocity properties of Danian limestone section exposed at the Curfs quarry, southeastern Netherlands. J. of Geol. And Min. Res. (2012); 4(4); 65-74. DOI: 105897/JGMR11.042.
- [10] Zijlstra JJP Brouwers MHMP Brinkuis H de Boer PL. Microfacies analysis of Cretaceous/Tertiary boundary sections. Geologie en Mijnbouw, 1996; 75: 133-155.
- [11] Radadi CM, Vanneau AA, Poupeau G, Carrio-Schaffauser E, Arnaud H, Rivera R. Interpretation of gamma-ray logs: the distribution of uranium in carbonates, Geosciences (2005); 337: 1457-1461.
- [12] Adams JA Weaver CE. Thorium to Uranium ratios as indicators of sedimentary processes: examples of concept of geochemical facies, Bull. Am. Assoc. Geol. (1956); 42, pp. 387-430.

To whom correspondence should be addressed: Oghenero Ohwoghere Asuma, Vrije Universiteit (VU) Amsterdam, Faculty of Earth and Life Science, De Boelelaan 1085, 1081 HVAmsterdam, The Netherlands, ohwonero@gmail.com