

Theoretical Establishment of the Mass Balance Equation and Determination of the Proportion $(S)_{ff}$ of Fossil Fuels as an Indicator of the Suess Effect

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Abstract

Mass balance equations can be used to quantify the level of fossil carbon dioxide $(CO_2)_{ff}$ at a given site by using samples of tree leaves or tree rings. These sites are often densely populated and highly industrialized areas. In this case, in addition to the natural carbon dioxide level of the site, there are several other components that are added to it. The approaches we propose in this paper are based on the isotopic fractionation of carbon and the laws of matter conservation. Mathematical equations are derived from this and we propose approaches to establish them. Our method has allowed us to come to the conclusion of the relationship that is already found in literature. But we term this relationship in another way by introducing the fraction of carbon modern F and the proportion of CO_2 derived from fossil fuels, which will be noted $(S)_{ff}$ for the following. As an indicator of the Suess Effect, we have calculated these values in our different sites. Wooded areas sites which are located in urban areas have higher value of proportion of fossil fuel $(S)_{ff}$.

Keywords

Suess Effect, Radiocarbon, Mass Balance, Fossil Fuel, $\Delta^{14}C$

1. Introduction

The conventional ^{14}C method, among its many applications, can also be used for

the determination of ^{14}C isotopic ratio values; $^{14}\text{C}/\text{C}$ ratio expressed in $\Delta^{14}\text{C}$ [1]. With this method one can get an idea of the degree of pollution and to be able to estimate CO_2 emissions from fossil fuels [1] [2] [3]. The estimation of fossil fuel CO_2 , the main cause of the Sues effect, cannot be determined directly from a radiocarbon age measurement [4] but can be estimated using a mathematical model for the global carbon cycle.

This discussion focuses on a proposed mathematical approach to establish the mass balance equations used by several authors in the determination of fossil CO_2 using the carbon-14 (C14 or ^{14}C) as a tracer.

The natural production of carbon-14 is generated by solar rays of cosmic and galactic sources. These solar rays contain protons that react with molecules in the air to release thermalized neutrons [5] with very high energy. These neutrons formed in the upper atmosphere then collide with air molecules such as nitrogen (preferentially) and oxygen. They are decelerated due to the many neutron-molecule collisions and attain the thermal energy of the gases. As a result, they react with the atomic nuclei of the air, present in the atmosphere and troposphere, to form carbon-14. The production of C14 is brought about by the capture of neutrons by nitrogen (80% in the atmosphere according to [6] is by far the most frequent reaction $^{14}\text{N} (n, p) ^{14}\text{C}$ [7].

2. Approach and Method

The properties of the same chemical element vary because of the difference in their masses. This difference is at the origin of isotopic fractionation, that is to say processes that vary the isotopic composition of organic or inorganic compounds.

For a given sample, its isotopic composition can be expressed either as the relative abundance of isotopes (in %); or according to the isotope ratio (without unit) or according to the isotope ratio in relation to a standard (noted δ and expressed ‰).

In this approach, we establish the equation of conservation of the isotopic composition of carbon in general and in particular C14 for a given sample of material.

The isotopic fractionation process can be described mathematically by comparing the isotopic ratios of the two equilibrium compounds ($a \leftrightarrow b$) or the two compounds before and after a physical or chemical transition process. For example the isotopic fractionation factor, named $\varepsilon_{a/b}$:

$$\varepsilon_{a/b} = \left(\frac{R_b}{R_a} - 1 \right) \quad (2.1)$$

R is an isotopic ratio. For example, the isotopic ratio of carbon can be defined by:

$$^{13}R = \frac{^{13}\text{C}}{^{12}\text{C}}, \text{ for the isotope } ^{13}\text{C} \quad (2.2)$$

$$\text{And } {}^{14}R = \frac{{}^{14}C}{{}^{12}C} \text{ for the } {}^{14}C \text{ isotope.} \quad (2.3)$$

It is the ratio of the least abundant and the most abundant isotope.

As far as literature is concerned, the three most abundant carbon isotopes are: ${}^{12}C$ (98.89%, the most abundant isotope), ${}^{13}C$ (1.1%) and ${}^{14}C$ (about 10^{-12} %).

Judging from the relations (2.2) and (2.3), the number of ${}^{12}C$, ${}^{13}C$ and ${}^{14}C$ isotopes in a sample is obtained respectively.

$${}^{12}C = \frac{C_G}{1 + {}^{13}R + {}^{14}R} \quad (2.4)$$

$${}^{13}C = \frac{{}^{13}RC_G}{1 + {}^{13}R + {}^{14}R} \quad (2.5)$$

$${}^{14}C = \frac{{}^{14}RC_G}{1 + {}^{13}R + {}^{14}R} \quad (2.6)$$

Since the compound has interacted with other sources of carbon, the carbon global (C_G) contained in one of the compounds is considered to come from several sources.

Each carbon source contributes to the overall carbon mass balance. Thus each source contains C_a , C_b and C_c etc. of carbon atoms of the same compound, a mass balance that allows to write for the global mixture:

$${}^{14}C_G = {}^{14}C_a + {}^{14}C_b + {}^{14}C_c + \dots \quad (2.7.a)$$

$${}^{13}C_G = {}^{13}C_a + {}^{13}C_b + {}^{13}C_c + \dots \quad (2.7.b)$$

$${}^{12}C_G = {}^{12}C_a + {}^{12}C_b + {}^{12}C_c + \dots \quad (2.7.c)$$

C_a , C_b and C_c etc. are the different components from the different sources that go into composition to form the carbon global mixture C_G in the sample.

Carbon reacts with atmospheric oxygen to give carbon dioxide CO_2 .

For a selected sample, for example, if it is a plant sample, or an oceanic sample etc. the amount of carbon dioxide measured $CO_{2\text{Sample}}$ is obtained with the contribution of at least three other CO_2 sources. There is the naturally (background) occurring source of $CO_{2\text{bg}}$, the fossil-fuel source $CO_{2\text{ff}}$ and other sources of biogenic origin, and unknown origin $CO_{2\text{other}}$.

We may write:

$${}^{12}CO_{2\text{Sample}} = {}^{12}CO_{2\text{bg}} + {}^{12}CO_{2\text{ff}} + {}^{12}CO_{2\text{other}} \quad (2.8.a)$$

$${}^{13}CO_{2\text{Sample}} = {}^{13}CO_{2\text{bg}} + {}^{13}CO_{2\text{ff}} + {}^{13}CO_{2\text{other}} \quad (2.8.b)$$

$${}^{14}CO_{2\text{Sample}} = {}^{14}CO_{2\text{bg}} + {}^{14}CO_{2\text{ff}} + {}^{14}CO_{2\text{other}} \quad (2.8.c)$$

By Liquid Scintillation Counting (LSC) or by accelerator mass spectrometry (AMS), we are interested in ${}^{14}C$. The ${}^{13}C$ and ${}^{12}C$ are stable isotopes, their composition does not vary.

(2.6) and (2.8.c) provide this relationship:

$$\frac{{}^{14}R_{\text{Sample}} \times CO_{2\text{Sample}}}{1 + {}^{13}R_{\text{Sample}} + {}^{14}R_{\text{Sample}}} = \frac{{}^{14}R_{\text{bg}} \times CO_{2\text{bg}}}{1 + {}^{13}R_{\text{bg}} + {}^{14}R_{\text{bg}}} + \frac{{}^{14}R_{\text{ff}} \times CO_{2\text{ff}}}{1 + {}^{13}R_{\text{ff}} + {}^{14}R_{\text{ff}}} + \frac{{}^{14}R_{\text{other}} \times CO_{2\text{other}}}{1 + {}^{13}R_{\text{other}} + {}^{14}R_{\text{other}}} \quad (2.9)$$

The isotopic abundances of carbon 13 on carbon 12 or carbon 14 on carbon 12 are very low in front of one. So they can be ignored in front of one.

Taking this approximation into account, relation (2.9) can produce the following equation:

$${}^{14}R_{Sample} \times CO_{2_{Sample}} = {}^{14}R_{bg} \times CO_{2_{bg}} + {}^{14}R_{ff} \times CO_{2_{ff}} + {}^{14}R_{other} \times CO_{2_{other}} \quad (2.10)$$

We specify that ${}^{12}CO_2$, ${}^{14}CO_2$, ${}^{13}CO_2$ are included in each of the following components: $CO_{2_{Sample}}$, $CO_{2_{bg}}$, $CO_{2_{ff}}$ and $CO_{2_{other}}$. It can be expressed as a relationship of this form:

$$CO_2 = {}^{12}CO_2 + {}^{13}CO_2 + {}^{14}CO_2 \quad (2.11)$$

According to [8] [9] [10], the isotope fractionation on carbon 13 is:

$$\delta^{13}C_S = 10^3 \left(\frac{{}^{13}R_{sample}}{R_{VPDB}} - 1 \right)$$

Then from that, we can have this:

$${}^{13}R_{sample} = R_{VPDB} \left(1 + \frac{\delta^{13}C_S}{10^3} \right) \quad (2.12)$$

We have VPDB = Vienna Pee Dee Belemnite which is used for the modern standard.

It should also be noted that we can well assume this approximation ${}^{14}R \approx ({}^{13}R)^2$.

Thus (2.10) become:

$$\begin{aligned} & {}^{14}CO_{2_{Sample}} \times \delta^{13}C_{Sample} \\ &= {}^{14}CO_{2_{bg}} \times \delta^{13}C_{bg} + {}^{14}CO_{2_{ff}} \times \delta^{13}C_{ff} + {}^{14}CO_{2_{other}} \times \delta^{13}C_{other} \end{aligned} \quad (2.13)$$

The fractionation for ${}^{14}C$ is almost exactly double that for ${}^{13}C$ [11] [12] so we can write:

$$\begin{aligned} & {}^{14}CO_{2_{Sample}} \times \delta^{14}C_{Sample} \\ &= {}^{14}CO_{2_{bg}} \times \delta^{14}C_{bg} + {}^{14}CO_{2_{ff}} \times \delta^{14}C_{ff} + {}^{14}CO_{2_{other}} \times \delta^{14}C_{other} \end{aligned} \quad (2.14)$$

If $\delta^{13}C = -25\%$, we have $\Delta^{14}C = \delta^{14}C$ and if not, we can write $\Delta^{14}C = \alpha\delta^{14}C + \beta$.

$$\text{(By this formula below: } \Delta^{14}C = \delta^{14}C - 2 \left(\frac{\delta^{14}C}{1000} + 1 \right) (25 + \delta^{13}C) \text{ [13])}$$

To the difference assumed or not introduced by the isotope fractionation on the sample, we can have:

$$\begin{aligned} & {}^{14}CO_{2_{Sample}} \times \Delta^{14}C_{Sample} \\ &= {}^{14}CO_{2_{bg}} \times \Delta^{14}C_{bg} + {}^{14}CO_{2_{ff}} \times \Delta^{14}C_{ff} + {}^{14}CO_{2_{other}} \times \Delta^{14}C_{other} \end{aligned} \quad (2.15)$$

3. Approach of Mass Balance Equation

For a sample of vegetable or an atmospheric sample, we can assume that the carbon dioxide CO_2 is formed:

- Of a component that represents the value of the CO₂ concentration in a certain level of the atmosphere (the troposphere) or background component free from any anthropogenic source. It is noted $(CO_2)_{bg}$.
- Of another component that represents the value of fossil fuel CO₂ concentration. This value is derived from the burning of fossil fuels. This is the fossil fuel CO₂ component. It is noted $(CO_2)_{ff}$.
- Of other large components that represent the value of the CO₂ concentration in the biosphere or exchanges between systems. It is noted $(CO_2)_{bio}$.
- And an obscure (obs) component that represents the value of unknown and uncontrolled sources of CO₂ concentration $(CO_2)_{obs}$.

These assumptions can be translated from the following equations:

$$(CO_2)_{Sample} = (CO_2)_{bg} + (CO_2)_{ff} + (CO_2)_{bio} + (CO_2)_{obs} \quad (3.1)$$

And

$$(CO_2)_{Sample} \times \Delta^{14}C_{Sample} = (CO_2)_{bg} \times \Delta^{14}C_{bg} + (CO_2)_{ff} \times \Delta^{14}C_{ff} + (CO_2)_{bio} \times \Delta^{14}C_{bio} + (CO_2)_{obs} \times \Delta^{14}C_{obs} \quad (3.2)$$

$(CO_2)_{Sample}$, $(CO_2)_{bg}$, $(CO_2)_{ff}$, $(CO_2)_{bio}$ and $(CO_2)_{obs}$ are respectively the global CO₂ concentration obtained from the sample, the background CO₂ concentration, fossil fuels CO₂ concentration, biogenic CO₂ concentration and unknown or/and uncontrolled (obscure) sources of CO₂ concentration.

$\Delta^{14}C_{Sample}$, $\Delta^{14}C_{bg}$, $\Delta^{14}C_{ff}$, $\Delta^{14}C_{bio}$ and $\Delta^{14}C_{obs}$ are respectively ¹⁴C level of the sample, the background ¹⁴C level, fossil fuels ¹⁴C level, sources of biogenic ¹⁴C level and unknown or/and uncontrolled (obscure) sources ¹⁴C level.

Finally, we will use these relations for the model of the equation that will allow us to find the local Suess effect caused by fossil fuels, as several authors such as [4] [14] [15] [16].

By (3.1) and (3.2) equations we can obtain the fossil-fuel CO₂ component $(CO_2)_{ff}$ that we can, without any approximation write in this way:

$$(CO_2)_{ff} = (CO_2)_{bio} \frac{\Delta^{14}C_{bio} - \Delta^{14}C_{Sample}}{\Delta^{14}C_{Sample} - \Delta^{14}C_{ff}} + (CO_2)_{bg} \frac{\Delta^{14}C_{bg} - \Delta^{14}C_{Sample}}{\Delta^{14}C_{Sample} - \Delta^{14}C_{ff}} + (CO_2)_{obs} \frac{\Delta^{14}C_{obs} - \Delta^{14}C_{Sample}}{\Delta^{14}C_{Sample} - \Delta^{14}C_{ff}} \quad (3.3)$$

To establish the mass balance equation, we will consider these approximations used by several authors like [4] [14] [16] [17] [18] [19] [20] and available in literature. These approximations make perfect sense.

1) Fossil carbon dioxide $(CO_2)_{ff}$ is free of significant amounts of radiocarbon, as fossil materials are several billion years old, a period of time long enough for the total amount of radiocarbon contained in these fossil organic materials to be removed.

Taking into account the relationship $\Delta^{14}C = 1000 \left(\frac{(^{14}C/C)_{Sample}}{(^{14}C/C)_{Std}} - 1 \right)$.

We can assume that $\Delta^{14}C_{ff} = -1000\text{‰}$ (3.a)

2) Other sources of carbon dioxide have their $\Delta^{14}C$ supposed equal to that of the background atmosphere. C-14 biogenic concentration $\Delta^{14}C_{bio}$ and C-14 obscure source concentration $\Delta^{14}C_{obs}$ are equal to the C-14 concentration in the background $\Delta^{14}C_{bg}$. (3.b)

Taking into account, these two approximations we can have:

$$(CO_2)_{ff} = (CO_2)_{bg} \frac{\Delta^{14}C_{bg} - \Delta^{14}C_{Sample}}{\Delta^{14}C_{bg} + 1000} \quad (3.4)$$

We will use this formula, in the following to determine the Suess effect of each sample used.

This formula is a universal one; no matter how many different components make up the overall carbon dioxide concentration measured in a sample $(CO_2)_{sample}$, as long as we assume that the ^{14}C concentration of the various components is taken to be equal to the background ^{14}C concentration.

If the nature of the samples used to quantify the fossil-fuel CO_2 concentration are not plants, this proposed methodology can be modified (oceanic samples for example).

To better quantify the contribution of fossil fuels to the level of carbon dioxide in the atmosphere, we will determine the proportion of CO_2 derived from fossil fuels, which will be called $(S)_{ff}$ for the following.

If we propose to work with samples, after 1950 (year zero in calendar ^{14}C), we can have this formula, available in the literature:

$$\Delta^{14}C = 1000 \left(F e^{-\mu(y-1950)} - 1 \right) \quad (3.5)$$

F is the fraction modern of component in the mixture; y is the year of sample collection and $\mu = \frac{\ln 2}{5568}$ is the radioactive constant decay of ^{14}C and 5568 years is the conventional half-life of Libby.

We want to use Equation (3.4) and (3.5) and then propose a new alternative equation that will be applicable regardless of the nature of the sample, vegetable etc.

(3.4) becomes:

$$(S)_{ff} (\%) = 100 \left(1 - \frac{F_{Sample}}{F_{bg}} \right) \quad (3.6)$$

The relation is independent of the time parameter.

F_{bg} and F_{Sample} is respectively the fraction of modern carbon for background and for the sample. $(S)_{ff} (\%)$ is the proportion of fossil-fuel derived CO_2 of the sample.

Now we will apply it to a particular case with results that we have already ob-

tained with the general method. At first, we give general information for our samples in **Table 1**.

Now we will tabulate the F-isotope fractionation values, in **Table 2**, that we found from the different samples we used. The method used is the same as we did in our previous work [21], *i.e.* Liquid Scintillation Counting (Code Laboratory DK) or by AMS-Arizona (Code Laboratory AA). By the way, due to the shortage of material available and more complete equipment, and in order to extend our measures, we have taken values from this article to have a continuous chronology until a more recent date.

In **Table 3**, we summarize the results of F_{bg} , F_{Sample} and $(S)_{ff}$. The proportion of fossil-fuel is calculated by the Formula (3.6).

$\Delta^{14}C_{bg}$ Values provide at [22] [23].

After calculating the proportion $(S)_{ff}$ of fossil fuels, to be seen in the **Table 3**, we have compared the values found:

- In wooded area: Mbao Forest (1960, 1966 and 2007) and UCAD Botanic Garden (1970, 1979, 1990, 2000).
- And in not wooded area often industrial sites: Highway (1961), SAR Factory (1964, 1968, 2005), Fass District (1976), Beach (1981, 1987, 1993, 1996, 1998 and 2009), Soumbédioune Market (2003) and Airport Runway (2010).

We notice that the values of the proportion of fossil fuels obtained in wooded area is more important that the values founded in not wooded area, with an average of 2.31% against 1.39%.

Sites where trees or botanical gardens grow and which are located in urban areas have higher value of proportion of fossil fuel $(S)_{ff}$. This is thought to be due to the enormous amount of anthropogenic CO_2 absorbed by the plants or trees.

But globally we can note, by **Figure 1**, that the values we found are small positive values. The largest value is 4.29% (in 1960 at wooded area) and the smallest value is 0.06% (in 1968 at not wooded area).

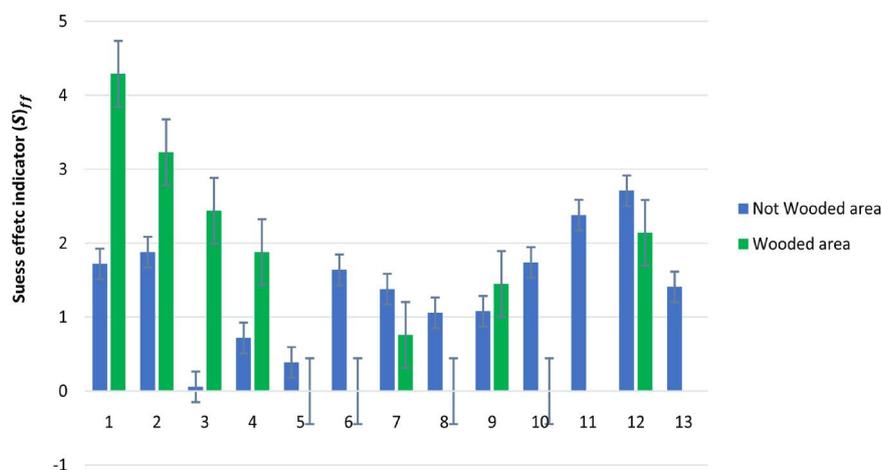


Figure 1. $(S)_{ff}$ values in wooded area and in not wooded area.

Table 1. General information for samples.

IFAN Herbarium reference	Nature	Year of Collect	Senegal Locality	Geographic Coordinates
29922	Herbaceous	1960	Mbao Forest 1	14°45'N, 17°18'W
04501	Herbaceous	1961	Highway	14°42'N, 17°26'W
53992	Herbaceous	1964	S.A.R Factory	14°44'N, 17°20'W
03414	Herbaceous	1966	Mbao Forest 2	14°46'N, 17°18'W
04730	Woody Species	1968	SAR Factory	14°44'N, 17°20'W
04738	Woody Species	1970	UCAD Botanic Garden 1	14°46'N, 17°27'W
20340	Woody Species	1976	Fass District	14°41'N, 17°27'W
54162	Herbaceous	1979	UCAD Botanic Garden 2	14°41'N, 17°28'W
04664	Herbaceous	1981	Beach 1	14°46'N, 17°23'W
04740	Herbaceous	1987	Beach 1	14°46'N, 17°23'W
04664	Woody Species	1990	UCAD Botanic Garden 3	14°41'N, 17°30'W
04741	Herbaceous	1993	Beach1	14°46'N, 17°23'W
04742	Herbaceous	1996	Beach 2	14°46'N, 17°23'W
54162	Herbaceous	1998	Beach 2	14°46'N, 17°23'W
28114	Woody Species	2000	UCAD Botanic Garden 4	14°41'N, 17°32'W
54163	Woody Species	2003	Soumbédioune Market	14°40'N, 17°27'W
54164	Herbaceous	2005	SAR Factory	14°44'N, 17°20'W
54165	Herbaceous	2007	Mbao Forest 1	14°45'N, 17°18'W
54166	Herbaceous	2009	Beach 2	14°46'N, 17°23'W
54167	Herbaceous	2010	Airport runway	14°44'N, 17°29'W

Table 2. F_{Sample} values in our samples.

Laboratory Code	IFAN Herbarium reference	Year of collect	$\delta^{13}C$	F_{Sample}
AA92997	29922	1960	-11.8	1.182 ± 0.005
AA92998	04501	1961	-29.4	1.202 ± 0.005
AA92999	53992	1964	-29.1	1.770 ± 0.008
AA93000	03414	1966	-28.2	1.674 ± 0.006
DK38	04730	1968	-25	1.600 ± 0.006
DK39	04738	1970	-25	1.518 ± 0.006
AA93001	20340	1976	-26.6	1.337 ± 0.005
DK40	54162	1979	-25	1.301 ± 0.005
AA9002	04664	1981	-25.5	1.272 ± 0.051
DK40B	04740	1987	-25	1.201 ± 0.005
DK41A	4739	1990	-25	1.181 ± 0.005
DK41B	04741	1993	-25	1.140 ± 0.005
DK41C	04742	1996	-25	1.121 ± 0.005
AA9003	28114	1998	-29.8	1.103 ± 0.006

Continued

DK42	54163	2000	-25	1.084 ± 0.006
AA9004	37650	2003	-23.7	1.073 ± 0.005
DK43	54164	2005	-25	1.064 ± 0.006
DK44	54165	2007	-25	1.052 ± 0.006
DK45	54166	2009	-25	1.041 ± 0.006
DK46	54167	2010	-25	1.050 ± 0.006

Table 3. Fossil fuel CO₂ results (in ppm).

Year (y) of collection	Background radiocarbon concentration ($\Delta^{14}C_{bg}$) in per mil	Background fraction of the modern carbon (F_{bg})	Sample fraction of the modern carbon (F_{Sample})	The Suess Effect Indicator values ($(S)_{ff}$) in %
1960	235	1.235	1.182 ± 0.005	4.29 ± 0.018
1961	223	1.223	1.202 ± 0.005	1.72 ± 0.007
1964	804	1.804	1.770 ± 0.008	1.88 ± 0.008
1966	730	1.73	1.674 ± 0.006	3.23 ± 0.011
1968	601	1.601	1.600 ± 0.006	0.06 ± 0.001
1970	556	1.556	1.518 ± 0.006	2.44 ± 0.009
1976	387	1.387	1.377 ± 0.005	0.72 ± 0.002
1979	326	1.326	1.301 ± 0.005	1.88 ± 0.007
1981	277	1.277	1.272 ± 0.005	0.39 ± 0.001
1987	221	1.221	1.201 ± 0.005	1.64 ± 0.007
1990	190	1.19	1.181 ± 0.005	0.76 ± 0.003
1993	156	1.156	1.140 ± 0.005	1.38 ± 0.006
1996	133	1.133	1.121 ± 0.005	1.06 ± 0.004
1998	115	1.115	1.103 ± 0.006	1.08 ± 0.005
2000	100	1.1	1.084 ± 0.006	1.45 ± 0.008
2003	92	1.092	1.073 ± 0.005	1.74 ± 0.008
2005	90	1.09	1.064 ± 0.006	2.38 ± 0.013
2007	75	1.075	1.052 ± 0.006	2.14 ± 0.012
2009	70	1.07	1.041 ± 0.006	2.71 ± 0.015
2010	65	1.065	1.050 ± 0.006	1.41 ± 0.008

4. Conclusion

At first, the objective of this paper is to exploit a simple method for establishing the mass balance equation. For the determination of the values of the fraction of modern carbon F of our samples, we use the ¹⁴C conventional method. Next, in order to estimate the Suess effect indicator, we have determined the fossil fuel fraction S_{ff} based on the mass balance equations for CO₂ concentration, stable isotopic carbon composition and ¹⁴C concentration. The results showed that our

values are positively small. (δ_{ff}) values in wooded area are larger than values in not wooded area.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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