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Determination of Radiocarbon concentration ($\Delta^{14}C$) and CO_2 emitted by fossil fuels in Dakar region (SENEGAL) from tree leaves using mass balance equations

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ABSTRACT: The aim of this work is to compare the local Suess effect in Dakar region to the global one during 20th century using tree leaves. Therefore, $\Delta^{14}C$ have been measured to give the variability in time. The curve obtained from this study compared to the global one used as reference is lower. The peak of curve for this study is 773‰ in 1964 where the one obtained in the same time by Nydal and Lovesth (1996) in Dakar is 800‰. These differences values are due to the local Suess effect corresponding to the emission of fossil fuel (CO_{2ff}) in the atmosphere. Fossil CO_2 (CO_{2foss}) is the major contributor of anthropogenic CO_2 to the atmosphere. In order to quantify these emissions the mass balance equations has been used allowing us to investigate the variability of the emissions. These analysis shows that CO_{2foss} decreases in wooded areas and increases in non-wooded areas, i.e. in more industrialized areas.

KEYWORDS: Suess effect, fossil fuel CO_2, $\Delta^{14}C$, radiocarbon.

1 INTRODUCTION

C-14 is a radionuclide the half-life of which is 5730±30 years. It is naturally produced in the atmosphere's upper layer from nuclear reactions between cosmic neutrons and air molecules ([**29**], **[5]**, **[28]**), particularly ^{14}N nitrogen. In addition to this production, we have anthropogenic origin of radiocarbon. First, there is the bomb effect (**[1], [6]**) which almost doubled the ^{14}C concentration (**[12]**, **[30], [31]**) and the Suess effect which led to a depletion of the ^{14}C concentration in the atmosphere (**[9]**,**[3]**,**[10]**,**[19]**, **[17]**,**[16]**, **[20]**,**[24]**, **[25]**, **[27]**, **[4]**).

Once it is formed, with other carbon isotopes (C-13 and C-12), C-14 is incorporated into the biological and geochemical cycles of carbon and is assimilated by all living organisms in the form of carbon dioxide after oxidation of this radio-isotope with air molecules like oxygen mainly. In any case, the concentration of $^{14}CO_2$ in the atmosphere can no longer be accepted as a constant, because the prevailing level of CO_2 will depend on the anthropogenic influence of modern times. So it has varied. Due to the high population demand for energy, the combustion of fossil fuels such as oil, gas, coal, etc. releases enormous quantities of CO_2 into the atmosphere. The consequence is a decrease in the carbon-14 concentration and an increase in the $^{14}CO_2$ concentration (**[19],[17],[16],[20],[24],[25],[27],[4]**) in the different carbon reservoirs. Today this phenomenon is much more caused by industrial complexes, the transport sector, the high human density in cities and other sources based on fossil fuel consumption.

Tree rings, leaves and short-lived plants (e. g. seasonal plants) assimilate carbon from the air during photosynthesis, so they better reflect changes in radiocarbon concentration (**[27]**) in the atmosphere and provide a good sample to better quantify the fossil fuel component of CO_2 in the atmosphere and the radiocarbon concentration.

In this investigation, the isotope ^{14}C is in use to estimate CO_2 in fossil fuel in an atmospheric sample. We first determined the radiocarbon concentration $\Delta^{14}C$ value of the samples in an attempt to better quantify the fossil contribution of CO_2 concentration in the atmosphere by using the mass balance equation.

2 SITES AND METHODS

2.1 SITES

Samples come from the Dakar region. We chose this locality, which is the capital of Senegal, because it is by far the most populated (23.2% of the country's total population and a density of around 5739 inhabitants per square kilometer) and the most industrialized (80% of the national sector) according to data from www.ands.sn.

The transport sector contributes significantly to air pollution in Senegal. Its gas emissions undoubtedly have serious impacts on urban air quality, particularly in Dakar and consequently on the health of populations later on. The concentration and poor condition of vehicles, due to their age, are sources of pollution. The vehicle fleet is ageing and constantly increasing.

Senegal is a country at the end of West Africa. It is bordered by the Atlantic Ocean to the West, Mauritania to the North, Mali to the East, Guinea and Guinée-Bissau to the South. Its geographical coordinates are: Latitude: 14° 41' 37'' North and Longitude: 17° 26' 38'' West.

This locality, which is densely populated, highly industrialized and with an accelerated growth of the land or air transport sector, is highly exposed to several forms of pollution such as PM5 and PM10, VOCs, methane, carbon monoxide (CO) and carbon dioxide CO_2 etc.

The focus of our study is on the contamination of air in the atmosphere by fossil CO_2 from fossil sources. Overall, we have selected five major sites (Table 1). The samples we treated are herbaceous or woody plants dating from 1960 to 2009.

We chose two types of sites: two wooded sites (UCAD Botanic Garden and Mbao Forest) and two other non-wooded sites (SAR Factory and Beach on the western cornice of Dakar).

Table 1. Sites, year of collection, geographical coordinates, species and laboratory code of samples

Sites	Year of collection	Geographical coordonates	Species	Code Labo
UCAD Botanic Garden	1970	14°41′N, 17°27′W	*Indigifera Costata*	DK39
	1979	14°41′N, 17°28′W	*Indigifera Costata*	DK40
	1990	14°41′N, 17°30′W	*Nérium Oleander*	DK41
	2000	14°41′N, 17°32′W	*Nérium Oleander*	DK42
Mbao Forest	1960	14°45 N, 17°18′W	*Cyperus Esculentus*	AA92997
	1966	14°46′N, 17°18′W	*Crotalaria Retusa Cyperus*	AA93000 DK44
	2007	14°45 N, 17°18′W	*Esculentus*	
SAR Factory	1964	14°44′N, 17°20′W	*Lactuca Aspera*	AA92999
	2005	14°44′N, 17°20′W	*Lactuca Intybacea*	DK43
Beach	1981	14°46′N, 17°23′W	*Indigofera Coptica*	AA93002
	1998	14°46′N, 17°23′W	*Ipomoca Coptica*	AA93003
	2009	14°46′N, 17°23′W	*Ipomoca Coptica*	DK45

2.2 METHODS

At the very beginning, our sample undergoes physical pre-treatment. The physical pre-treatments have been made to eliminate all physical impurities present in the sample before carrying out the chemical treatments. Physical pretreatment is generally a manual process that is often less complicated.

The chemical pretreatments that we performed for our samples are routine acid-base-acid. The samples were washed in distilled water and treated with hydrochloric acid solution 0.5 M and then these samples were rinsed with distilled water. Then we make an attack with a NaOH sodium hydroxide solution 0.1M. After about one hour, we rinse the samples with distilled water and then make a new acid attack. Then, the samples were washed again with distilled water and dried in a dryer at a temperature of about 30°C.

Then we progress to the stage of benzene synthesis. Benzene synthesis consists of three parts: the sample carburizing step, acetylene synthesis and acetylene trimerization to obtain benzene.

This entire reaction chain is carried out on a synthesis bath of benzene in use in our laboratory. Then we move on to the counting stage. We use a conventional method with a liquid scintillation counter (Tricarb 3170TR / SL) with a super low level option. In order to minimize background interference and discriminate against legitimate beta events (β), the Tricarb 3170TR / SL is equipped with a bismuth and germanium detector as well as a peak analyzer.

Standardization is performed routinely to see the electronic stability of the counting system. The optimization of the counting zone has been done to maximize the merit factor (E^2 / B) where E is the counting efficiency and B is the background counting rate, this type of counter is equipped with a SNC (Self Normalization and Calibration) cassette. These samples are coded DKxx. Some parts of our samples were measured by Accelerator Mass Spectrometry by the NSF-Arizona Physics Laboratory in the United States. These samples are coded AAxx Arizona AMS).

3 RESULTS AND DISCUSSION

Then we discuss the radiocarbon measurement results obtained from our measurement sites. For $\Delta^{14}C_{mes}$ values we have use the formula $\Delta^{14}C = \left(Fe^{\lambda(1950-y)} - 1\right)1000$ (before 1950) or $\Delta^{14}C = \left[Fe^{\lambda(y-1950)} - 1\right]$ (after 1950). And Fraction of modern carbon $F^{14}C$ of sample are obtained according to $F^{14}C = {A_{SN}}/{A_{STD}}$.

In the following table (**Table2**) we show the values of $\Delta^{14}C_{mes}$ and $\Delta^{14}C_{bg}$. For $\Delta^{14}C_{bg}$, we have choosen Mauna Loa (MLO) as background, in order to compare the radiocarbon concentration of sample and that of clean area.

We chose the measurement data from the Mauna Loa station in Hawaii (United States of America) (Globalview-CO_2-2010) because this site according to **[2]**, is in the same hemisphere as Senegal (NH Zone 2). Countries in the same hemisphere have approximately the same level of background. And moreover, we take $\delta^{13}C$= -25 ‰ for our samples.

Table 2. Modern Fraction of Carbon F, $\Delta^{14}C_{mes}$ and $\Delta^{14}C_{bg}$ per year

Samples	**Year**	**F**	**$\Delta^{14}C_{mes}$ (‰)**	**$\Delta^{14}C_{bg}$ (‰)**
UCAD Botanic Garden	1970	1,518±0,005	522±6	556
	1979	1,301±0,005	306±5	326
	1990	1,181±0,005	187±5	190
	2000	1,084±0,006	91±6	100
Mbao Forest	1960	1,182±0,005	184±5	235
	1966	1,674±0,006	677±5	735
	2007	1,052±0,005	60±6	75
SAR Factory	1964	1,770±0,008	773±5	800
	2005	1,07±0,006	71±6	90
Beach	1981	1,272±0,051	277±5	277
	1998	1,103±0,006	110±5	115
	2009	1,049±0,005	49±6	70

In the following figure we compare the concentration of radiocarbon obtained from our samples with its concentration in a clean air environment (Background).

We have represented these two curves of variations in the same graph.

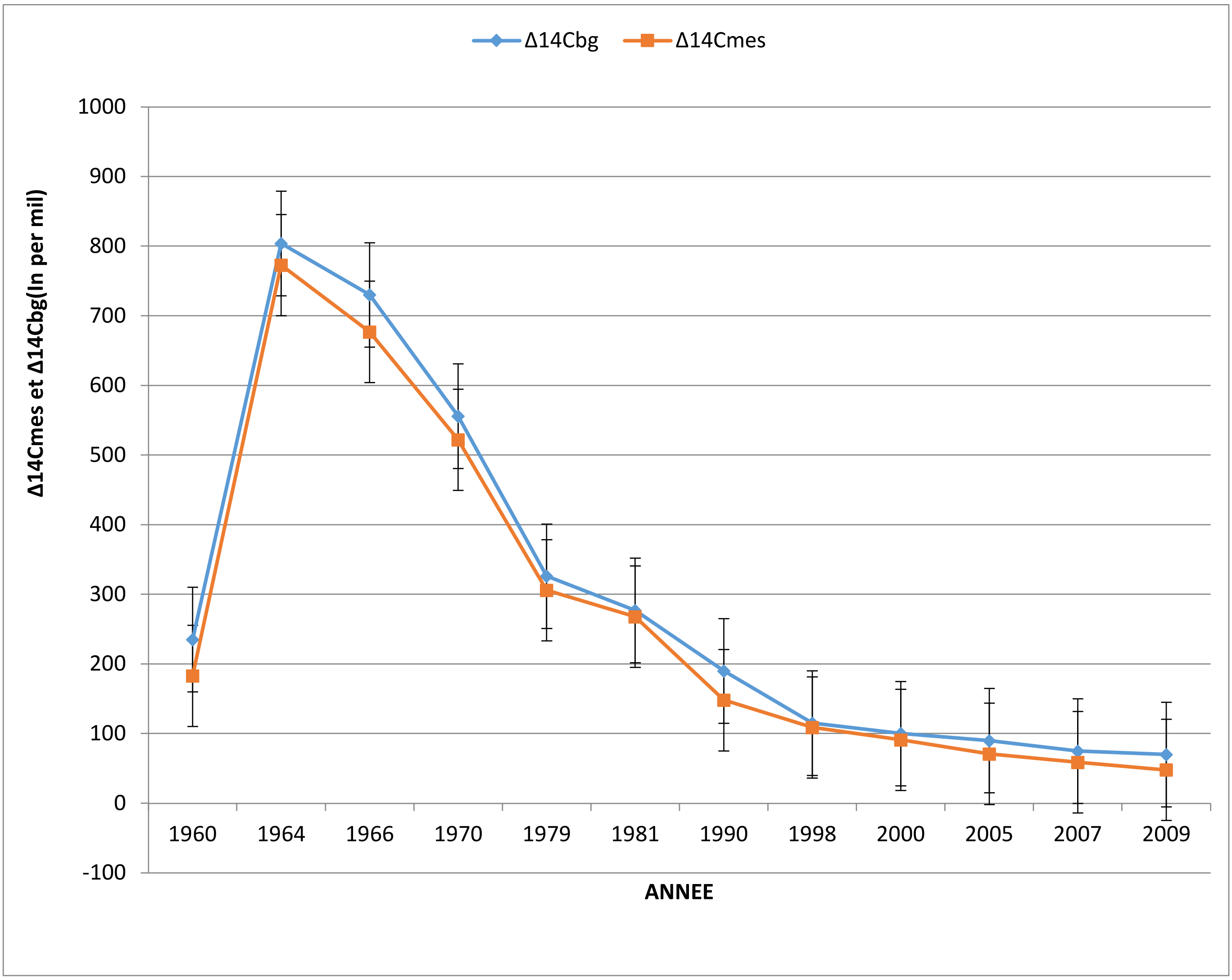

Fig. 1. Comparison of concentrations $\Delta^{14}C_{mes}$ and $\Delta^{14}C_{bg}$. Values are in per mil.

Figure 2 shows the variations of $\Delta^{14}C$ in tree leaves and the variations of $\Delta^{14}C$ in a clean air environment between 1960 and 2009. There is a difference between the values of the measured radiocarbon concentration $\Delta^{14}C_{mes}$ and those of the ^{14}C concentration $\Delta^{14}C_{bg}$ in a clean zone.

Our graph consists of two parts:

- From 1958 to around 1964, corresponding to the time the bombs occurred when the values of $\Delta^{14}C$ increased regularly until they reached a value of 773±5‰. This increase in the level of $\Delta^{14}C$ is due to Nuclear tests and weapons. (**[8]**, **[4]**)
- From 1968 to 2009, there was a decrease in the ^{14}C level until 70‰.This drop in level is due to fossil fuels used by man (**[27]**,**[6]**, **[20]**,**[4]**).

Atmospheric data from other sites (Nydal and Lovseth 1996) show a peak of 817±23 ‰ in 1964.Compare to our results, we have 773±8‰ in the same date. According to this graph, it can be noted that the concentration of ^{14}C in "clean" air is generally higher than the concentration of ^{14}C in an environment supposed to be a "polluted" area given in Dakar. The difference is due to the Suess effect.

We will now determine the fossil CO_2 concentration from the mass balan equation.

Now lets remind the mass balance equation.

From a quantitative and material conservation, for a given material sample, we can assume these following relations:

$$[CO_2]_{total} = [CO_2]_{bg} + [CO_2]_{bio} + [CO_2]_{foss} \quad \textbf{(1)}$$

$$[CO2]_{total}\,\Delta^{14}C_{total} = [CO2]_{bg}\,\Delta^{14}C_{bg} + [CO2]_{bio}\,\Delta^{14}C_{bio} + [CO2]_{foss}\,\Delta^{14}C_{foss} \quad \textbf{(2)}$$

To design the model for the equation that will allow us to find the local Suess effect caused by fossil fuels, as several authors such as ([**12**], [**18**], **[2]**) have done, we have combined the relationships **(1)** and **(2).**

By substituting equations **(1)** and **(2)** we can obtain the fossil CO_2 component (CO_{2foss}) that we can, without any approximation write in this way :

$$[CO_2]_{foss} = [CO_2]_{bio}\frac{\Delta^{14}C_{bio} - \Delta^{14}C_{total}}{\Delta^{14}C_{total} - \Delta^{14}C_{foss}} + [CO_2]_{bg}\frac{\Delta^{14}C_{bg} - \Delta^{14}C_{total}}{\Delta^{14}C_{total} - \Delta^{14}C_{foss}} \quad \textbf{(3)}$$

To establish the mass balance equation, we will consider these approximations used by several authors like **([12]**, **[14]**, **[12]**, **[7]**, **[32]**, **[18]**, **[25]**) and available in the literature. These approximations make perfect sense.

a) Fossil carbon dioxide CO2foss 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. So we can assume that $\Delta^{14}C_{foss} = -1000‰$

b) C-14 biogenic concentration $\Delta^{14}C_{bio}$ is equal to the C-14 concentration in the background $\Delta^{14}C_{bg}$ because the main flow of the biosphere comes from autotrophic respiration (it has not been affected by fossil fuel contribution) and we can write: ($\Delta^{14}C_{bio} = \Delta^{14}C_{bg}$)

Taking into account, these two approximations we can have:

$$\mathrm{CO}\,2_{foss} = CO_{2mes}\,\frac{(\Delta^{14}C_{bg} - \Delta^{14}C_{mes})}{\Delta^{14}C_{bg} + 10^{3}} \quad \textbf{(4)}$$

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

The results obtained for our different samples are recorded in the following table:

Table 3. Sites, year of sample, and fossil CO_2 value of the sites

Samples	Year	CO_{2mes} (ppm)	$\Delta^{14}C_{bg}$ (‰)	$\Delta^{14}C_{site}$ (‰)	CO_{2ff} (ppm)
UCAD Botanic Garden	1970	325,68	556	522	7,1
	1979	336,84	326	306	5,1
	1990	354,39	190	187	2,7
	2000	369,55	100	91	3
Mbao Forest	1960	316,91	235	184	13,3
	1966	321,28	735	677	10,7
	2007	383,79	75	60	5,3
SAR Factory	1964	319,62	804	773	5,5
	2005	379,8	90	71	6,6
Beach	1981	340,11	277	268	2,4
	1998	366,7	115	110	1,9
	2009	387,43	70	49	7,6

Radiocarbon (^{14}C) in atmospheric CO_2 in the Dakar urban area (Senegal) was measured using tree leaves. The first objective of our study was to evaluate the local Suess effect in Dakar and to compare the concentration of ^{14}C measured in a sample of tree leaves with that of ^{14}C in a "clean" air environment.

Then in a second step, after determining the fossil CO$_2$ concentration, we compared our results with those of the World Bank data on fossil CO$_2$ emissions (in metric tons per capita) in Senegal during the same period from 1960 to 2009.

1. Carbon dioxide emissions are emissions from the combustion of fossil fuels and the manufacture of cement. They include carbon dioxide emissions produced during the consumption of solid, liquid or gaseous fuels and flaring.
2. The values of fossil CO$_2$ emissions (in metric tons per capita) in Senegal come from the Centre d'analyse des informations relatives au dioxyde de carbone, division des Sciences de l'environnement, Oak Ridge National Laboratory, Tennessee, USA.

Table 4. CO$_2$ fossile (in ppm) and CO$_2$ (in metric tons per capita)

ANNEE	CO$_{2foss}$ (in ppm)	CO$_2$ (in metric tons per capita)
1960	13,3	0,258
1964	5,5	0,258
1966	10,7	0,454
1970	7,1	0,307
1979	5,1	0,533
1981	2,4	0,574
1990	2,7	0,423
1998	1,9	0,367
2000	3	0,402
2005	6,6	0,524
2007	5,3	0,443
2009	7,6	0,372

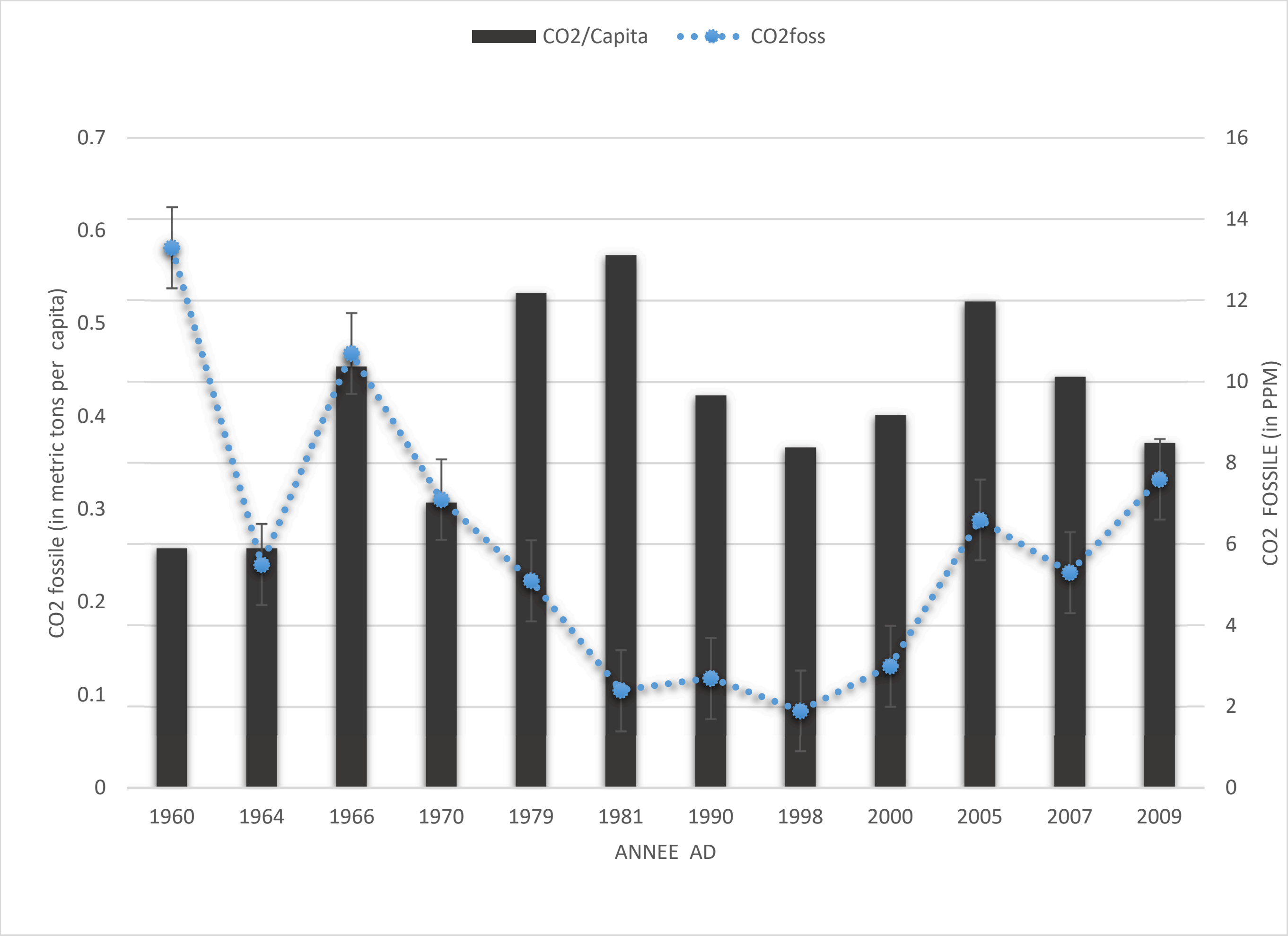

Fig. 2. ***Comparison of fossil CO_{2ff} values (in ppm) from our samples and fossil CO_2 values (in metric tons per capita) from 1960 to 2009***

Below is the graph of the formal comparison of the fossil CO_2 values of our samples and the CO_2 values in metric tons per inhabitant relative to the World Bank data.

From a form standpoint, both curves look the same.

As the quantities issued do not have the same unit, it will be possible to compare them from a formal point of view.

[10], [**18]**, **[19]**, **[16]**, **[24]**, **[27]**, **[20]**, **[4]** have shown this phenomenon through different studies at different times. Today, this phenomenon is caused, most often, by industrial complexes, the transport sector, the high human density in the cities and other sources based on the consumption of fossil fuel.

In order to show the reliability of our results, we have compared our results, namely the comparison of the point of shape and shape, of the curve of variation of fossil CO_2 emissions per inhabitant with that of the World Bank data concerning fossil CO_2 emissions (in metric tons per inhabitant) of Senegal during the same period from 1960 to 2009.

It can be said that the overall variations in CO_2 emissions from calculated fossil fuels are consistent with those of the World Bank data.

However, there are a few moments when we observe a reduction of fossil CO_2 in our calculations while we notice the reverse phenomenon at the level of data from the World Bank. This difference is explained by the fact that our values concern fossil emissions of specific points in the Dakar region, where as the World Bank data concern the global averages of Senegal. It may also be noted that the influence of the different methods of measurement and their units of values which are not the same, is a non-negligible factor.

If we had measurements of CO_2 emissions per metric ton per inhabitant, only in the Dakar region, the calculated fossil CO_2 values would be closer to the measured fossil CO_2 values.

4 CONCLUSION

The objective of this work was to implement the radiocarbon dating method to measure fossil CO_2 emissions in urban areas of Senegal.

To do this quantification we used the radiocarbon dating method also using a TriCarb 3170/R liquid scintillation counter in super Low Level mode. The samples we worked with were tree leaves because these plants better reflect the contamination of "pure" air by CO_2 from the atmosphere.

The concentrations of fossil CO2 (CO_{2foss}) found in our samples are in agreement with those available in the datawordbank.org database of the World Bank for Senegal. Although the units are not the same, parts per million (ppm) for our results and metric tons for World Bank values.

The results found show a decrease in radiocarbon concentration and an increasing increase in the amount of CO2 in the atmosphere.

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