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RESEARCH ARTICLE

Thermoelectrical power plant influence on environmental radiocarbon level in the Govora industrial area

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Abstract

The radiocarbon (^{14}C) specific activity was measured in vegetation and atmosphere in Ramnicu Valcea, Romania. On the sampling location operates a nuclear installation, namely "Experimental Pilot for Separation of Tritium and Deuterium" (PESTD), a semi-industrial installation designed for the detritiation of heavy water moderator of CANDU reactors and a 315 MW coal-fired thermoelectric power plant. Because one of the important releases of PESTD is gaseous radioactive effluent, the baseline of radiocarbon was a must for the environmental program. On the other hand, due to the Suess effect, a relative decrease of the radiocarbon-specific activity on a local scale is expected as a result of the dilution of the carbon isotopic mixture by fossil carbon. All the measurements were done by liquid scintillation counting and direct absorption method. It can be observed that the specific activity of ^{14}C was similar for both types of samples investigated. The variations encountered are generally within the limit of uncertainty associated with the ^{14}C. The average radiocarbon-specific activity recorded has the following values: 0.226 ± 0.016 Bq/gC for the vegetation and 0.228 ± 0.016 Bq/gC for the atmosphere. The results have a clear decreasing trend, but due to local influence caused by the continuous production of fossil CO_2, cannot be observed ^{14}C seasonal variations. A strong correlation between radiocarbon activity in air and vegetation was highlighted.

Introduction

Radiocarbon (^{14}C) is a common radionuclide in the environment. It is the only radioactive isotope of carbon with a long half-life, 5700 ± 30 years (LNHB). Natural production of radiocarbon takes place at high altitudes in the atmosphere due to the nuclear reaction of thermal neutrons, generated by cosmic rays, with ^{14}N nuclei (IRSN). ^{14}C thus oxidizes to carbon dioxide and results in a flux of $^{14}CO_2$ in the troposphere where it is incorporated into plants through photosynthesis, as well as in the meteoric and ocean waters through CO_2 exchange reactions. This makes ^{14}C an ideal tracer of carbon dioxide coming from the combustion of fossil fuels (Rakowski 2011; Zazzeri 2023) or from other anthropogenic activities such as the nuclear industry (Jean-Baptiste 2018; Kontul 2018).

Burning of fossil fuels causes the emission of carbon dioxide into the atmosphere. The isotopic fingerprint of CO_2 from fossil sources is slightly different from that from modern sources, mostly because of the very long formation period of fossil fuels, which therefore do not contain radiocarbon. This was first noted in the mid-1950s by the Austrian chemist Hans Suess (Suess 1955) who proved that contemporary tree-sample radiocarbon activity was lower than in samples from the middle of the 19th century before the Industrial Revolution, based on fossil fuels (coal, natural gas, petroleum). This is called the Suess effect (Keeling 2017; Sonnerup 1999) and it is defined as a change in the ratio of the atmospheric concentrations of heavy isotopes of carbon (^{13}C and ^{14}C) by the admixture of fossil-fuel-derived CO_2, which is depleted in $^{13}CO_2$ and contains no $^{14}CO_2$ (Tans 1979). There are studies in the literature that have shown that atmospheric $\Delta^{14}C$ (Stuiver 1977) decreased by about 20.0‰ between the

years 1890 and 1950 from which 17.3‰ is attributed to the Suess effect and the rest is attributed to the variation in the cosmic ray production of radiocarbon (Stuiver 1981). Even if 95% of the total carbon dioxide from the burning of fossil fuels is produced in the Northern Hemisphere due to air-mass mixing between the Northern and Southern Hemispheres, the Suess effect has a global character.

According to the literature, nuclear weapons testing in the 1950s and 1960s nearly doubled the atmospheric radiocarbon. The level of bomb radiocarbon was about 100% above normal levels between 1963 and 1965. The level of bomb radiocarbon in the Northern Hemisphere reached a peak in 1963, and in the Southern Hemisphere around 1965 (Currie et al. 2011; Hua and Barbetti 2007; Levin and Kromer 2004; Levin et al. 2010). Radiocarbon releases from nuclear facilities are of great interest because radiocarbon is the most important radionuclide from the point of view of the radiation doses received by the population due to the nuclear industry (UNSCEAR 2017). The radiocarbon produced by nuclear facilities is released mostly in the form of gaseous discharges (IAEA 2004). Currently there is no satisfactory technology for removing radiocarbon from effluents and its discharge, therefore radiocarbon releases should be monitored.

The results presented in this paper represent radiocarbon activity in atmospheric carbon dioxide and evergreen plants in Ramnicu Valcea, Romania. The aim has been to study the influence of a thermoelectrical power plant on radiocarbon variation in the vegetation and atmosphere of the Govora Industrial Area. This study is the continuation of the one previously carried out by the authors (Faurescu 2019) in which the radiocarbon level from the same location was compared with $^{14}CO_2$ observations at Jungfraujoch, a high-altitude research station in the Swiss Alps conducted to monitor the background $^{14}CO_2$ level over Europe and to define the reference for regional estimates of fossil fuel CO_2 (Hammer 2017).

Materials and methods

The sampling was done in the Govora industrial area placed about 10 km south of Ramnicu Valcea city, Romania (Figure 1). This location has some particularities. The first particularity to take into account is the fact that near the sampling location operates a nuclear installation, namely "Experimental Pilot for Separation of Tritium and Deuterium" (PESTD) designed for the detritiation of heavy water moderator of CANDU reactors. Until now, PESTD's normal operation was with heavy water and tritiated water below the exemption level approved by Romanian legislation. Heavy water reactors emit significant amounts of tritiated water and ^{14}C, the ^{14}C being a by-product resulting primarily from neutron activation of ^{17}O from heavy water molecules. Because one of the important releases of PESTD is gaseous radioactive effluent, the monitoring of atmospheric ^{14}C was included in the environmental monitoring program.

Another particularity is that the Govora industrial area operates a 315 MW coal-fired thermoelectric power plant. Due to the Suess effect, a relative decrease of the ^{14}C activity on a local scale is expected as a result of the dilution of the carbon isotopic mixture by fossil carbon. To determine the thermoelectrical power plant's influence on environmental radiocarbon levels in the Govora industrial area, the radiocarbon activity in the atmosphere and vegetation was determined by liquid scintillation counting and the direct absorption method. The samples were collected for two years as follows: air samples collected every two weeks by active absorption of CO_2 into sodium hydroxide (NaOH 3M) with a Raschig tube, and monthly evergreen vegetation (*Thuja occidentalis L.*) samples.

For the air samples, the sampling device used was made in our laboratory and it is similar to the one developed by the Central Radiocarbon Laboratory affiliated with the Institute of Environmental Physics of Heidelberg University (Hammer 2017). To ensure a fast and reliable method for radiocarbon measurements in air the schematic set-up of the sampler (Figure 2) is as follows.

The air is pumped by an air pump with variable flow and passed through an air flow meter that allows adjusting the flow of pumped air. To have enough CO_2 to saturate the liquid scintillation cocktail as described below, a sampled air volume of approximately 25 m^3 is required. Therefore, for a sampling

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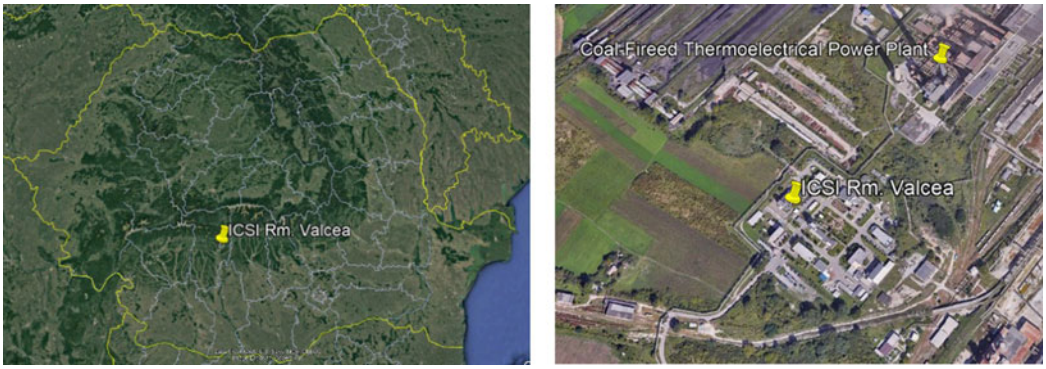

Figure 1. *Location of the sampling point in the Govora industrial area, Romania (adapted from Google Earth).*

Figure 2. *The schematic set-up of the atmospheric CO_2 sampler by active absorption.*

Figure 3. *(a) Raschig tube and (b) glass Raschig rings.*

time of two weeks, this volume of air is reached if air is sampled with a flow rate of 75 L/hr. After the flow meter, the air passes through a gas meter where the sampled air volume is monitored. Then, the air passes through a gas-washing bottle to be humidified. This prevents the NaOH solution from being too concentrated and solid deposits of sodium carbonate in the Raschig tube (Figure 3). The gas-washing bottle is connected to the Raschig glass tube, where the atmospheric CO_2 is absorbed by the NaOH solution. The tube is rotated slowly by a motor so that the NaOH solution will be permanently renewed on the Raschig rings. To provide a large surface area within the volume of the tube for interaction between liquid and gas were used 6 mm × 6 mm × 1 mm borosilicate glass Raschig rings (Lenz Laborglas GmbH, Cat. No. 5 1270 06).

For vegetation samples, evergreen vegetation samples were chosen i.e. thuja leaves (*Thuja occidentalis L.*). Evergreen vegetation can photosynthesize through the winter as long as they are not frozen and have access to water (Oquist 2003). Although maximum photosynthetic rates of evergreen species decrease in winter, the photosynthetic ability is still maintained (Miyazawa 2005), and CO_2 assimilation takes place. Since Romania has a temperate continental climate, the mean annual temperature is 11°C in the south of the country and 8°C in the north of the country (MEW 2005). Even

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so, the mean temperature in the winter period falls below –3°C, but below-zero periods are short, and evergreen plants' photosynthesis is stopped only in these periods.

Radiocarbon measurements were done by direct absorption method and liquid scintillation counting (Leaney 1994; Varlam 2006). The method involves capturing carbon dioxide from the sample in the scintillation cocktail as carbamate and measuring it in an ultra-low level liquid scintillation spectrometer Quantulus™ 1220. The scintillation cocktail is homemade and contains CarbonTrap (Meridian Biotechnologies Ltd), fluorescence substances PPO and bis-MSB (PerkinElmer), and solvents methanol and toluene. Depending on the type of sample, carbon dioxide is obtained either by combustion as in the case of vegetation samples, or by acidification with HCl in the case of the NaOH solution in which atmospheric CO_2 was captured. Before combustion, vegetation samples were dried to constant weight at 60ºC, ground, and 10 g of sample was pelletized. Combustion was done in an oxygen atmosphere (17 atm.) in a Parr 1121 combustion vessel (Moghissi 1975). It can accommodate samples weighing up to 10 grams using oxygen charging pressures up to 20 atm. The vessel is useful for determining trace amounts of tritium, carbon-14, or heavy metals in vegetable matter. In the case of sodium hydroxide solution, pure CO_2 was obtained by acidification with HCl, and the pure CO_2 was collected in a gas bag (Supelco, product no. 24655). The direct absorption of CO_2 into the scintillation cocktail was done by bubbling according to the set-up described in the literature (Faurescu 2019). This setup contains a pump for gas vehiculation, two purification traps containing an aqueous solution of $AgNO_3$ to retain water vapors and chloride, a flowmeter, and a bubbler with the scintillation cocktail. Gas bubbling for 10 min with a flow rate of about 0.2 L/min through the scintillation cocktail was enough to ensure saturation of amine with the CO_2 as carbamate.

Next, the amount of CO_2 captured in the scintillation cocktail is determined by weighing. The mass of CO_2 is the weight difference between the bubbler with the scintillation cocktail before and after bubbling. For the measurement by liquid scintillation counting method, the entire scintillation cocktail was transferred in 20-mL low-potassium glass vials (PerkinElmer, product no. 6000128) and then counted using an ultra low-level liquid scintillation spectrometer, Quantulus 1220. The counting efficiency was established with the internal method. This method involves the measurement of a standard sample together with a background sample. The background sample was prepared in a similar way as the unknown samples with CO_2 obtained by acidification with HCl from marble. Marble was chosen for a background because it does not contain radiocarbon due to the very long time it takes to form. The standard sample is a background sample in which a standard capsule with known activity (PerkinElmer, product no. 1210-122) was dissolved. The standard sample, the background sample, and the unknown samples were counted for 10 cycles of 100 minutes (1000 min counting time). The counting efficiency at the best factor of merit was around 65% with a background of around 2.3 CPM (counts per minute). A double check of the CO_2 saturation of the scintillation cocktail was done by checking the spectral quench parameter of the external standard, SQP€. We obtained the same SQP€ values around 630 ± 1%. Data acquisition was performed by using WinQ Windows workstation software, and for spectra processing 1224-534 EASY View software was used.

Results and discussion

Validation of LSC radiocarbon measurement of environmental samples was performed through an intercomparison exercise. The ^{14}C concentration were determined on five environmental samples (thuja leaves, *Thuja occidentalis L.*) by liquid scintillation counting and accelerator mass spectrometry (AMS) (Table 1). LSC measurements were performed at the National R&D Institute for Cryogenic and Isotopic Technologies – ICSI Rm. Valcea and AMS measurements at the Horia Hulubei National Institute for R&D in Physics and Nuclear Engineering (IFIN-HH). The results of the radiocarbon measurements are reported in pMC according to Stuiver and Polach (Stuiver 1977). The $\delta^{13}C$ ratio was measured by isotope

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Table 1. *Results obtained from the intercomparison exercise LSC–AMS*

Sample	$\Delta^{13}C$ (‰)	A_{LSC} (pMC)	U_{LSC} (pMC)	A_{AMS} (pMC)	U_{AMS} (pMC)	ζ
1	–28.06	102.5	3.8	101.0	1.0	–0.46
2	–27.36	103.4	3.7	102.1	1.0	–0.34
3	–29.20	102.5	3.7	102.3	1.0	–0.05
4	–28.52	106.6	3.8	103.6	1.0	–0.73
5	–27.40	103.0	3.8	101.5	1.0	–0.38

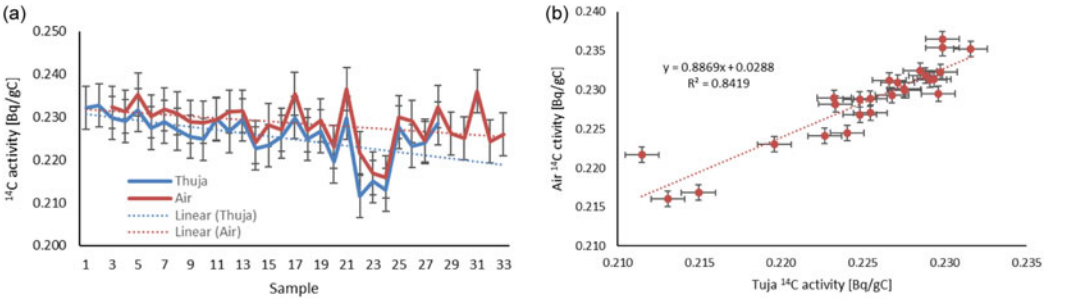

Figure 4. *Variation of radiocarbon activity in air and vegetation during (a) the observation period, and (b) the correlation between radiocarbon activity in air and vegetation.*

ratio mass spectrometry on a Delta V IRMS on dried thuja leaves samples. The comparison of results was performed with the ζ-test calculated according to the following equation:

$$\zeta = \frac{A_{AMS} - A_{LSC}}{\sqrt{U_{AMS}^2 + U_{LSC}^2}}$$

where A_{AMS} is activity reported by ASM lab and A_{LSC} activity reported by LSC lab and U_{AMS} and U_{LSC} are the appropriate combined uncertainties with coverage factor k=1. Results for which the absolute values of the ζ-test are less than 1.64 are in agreement. Results between 2.56 and 1.64 are questionable.

The results were satisfactory and showed that radiocarbon measurements by LSC provide reliable values although the LSC uncertainties are much higher than that of the AMS measurements.

To establish the influence of the thermoelectrical power plant on environmental radiocarbon levels in the Govora Industrial Area we measured the radiocarbon activity in the atmosphere and the thuja leaves sampled from this location.

The results have a decreasing trend, but due to local influence caused by the continuous production of fossil CO_2, we cannot observe a seasonal variation. For the radiocarbon activity in the air samples, the minimum value recorded was 0.211 ± 0.014 Bq/gC in a winter month, the maximum value of 0.233 ± 0.016 Bq/gC in a summer month, and an average value of 0.226 ± 0.016 Bq/gC. Similar values were recorded for thuja leaves, sampled in the same period as air samples, with a minimum of 0.216 ± 0.014 Bq/gC, a maximum of 0.237 ± 0.018 Bq/gC, and an average of 0.228 ± 0.016 Bq/gC. The Pearson correlation coefficient between the radiocarbon activity in air and the vegetation was 0.92 which suggests a strong correlation between them. The R^2 value of 0.842 indicates also a pretty good fit of the estimated trendline values to the measured values (Figure 4, b).

Despite the good correlation between the radiocarbon activity in the atmosphere and thuja leaves sampled in the same period, we wanted to see if the same correlation is maintained for a delay in the assimilation of the radiocarbon concentration by the vegetation for one month, two months or three

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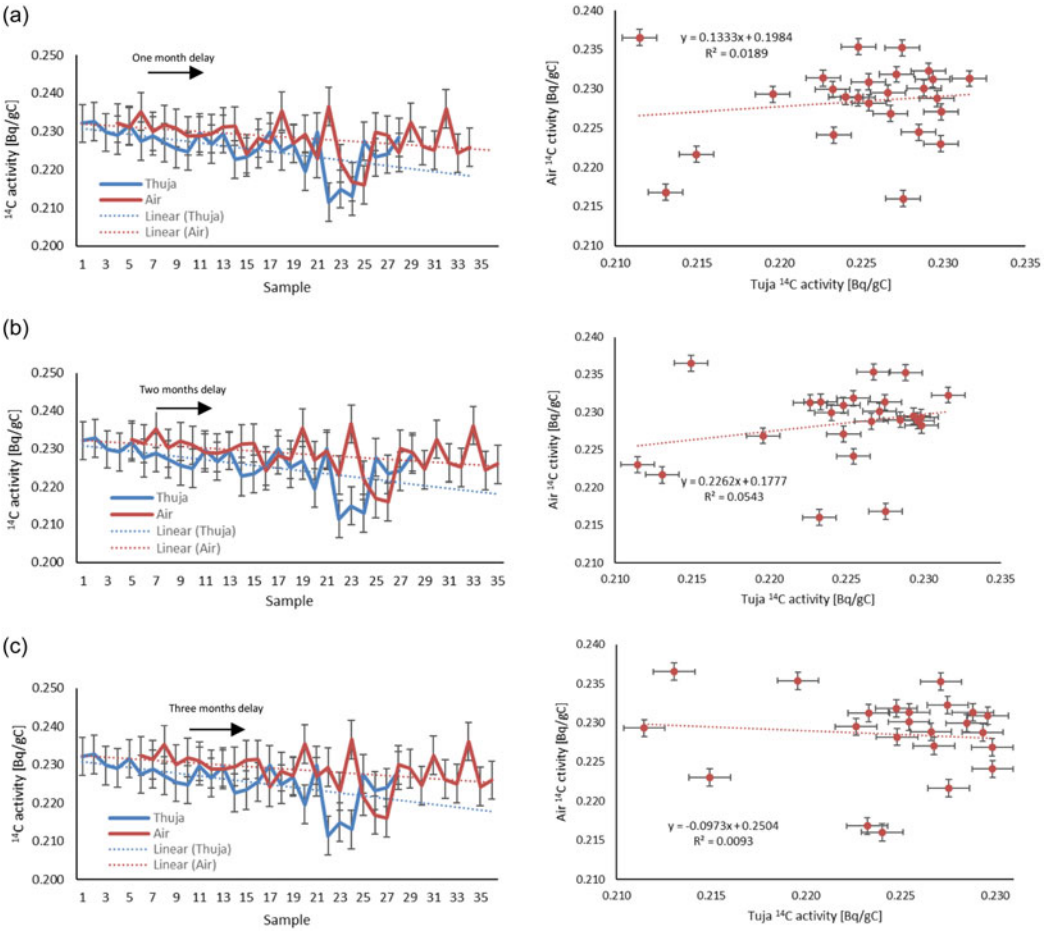

Figure 5. *The correlation between radiocarbon activity in air and vegetation: (a) one month delay, (b) two months delay, and (c) and three months delay.*

months. Figure 5 shows the correlations between radiocarbon activity in air and vegetation in the three scenarios.

It can be observed that the correlation between the two parameters is getting worse as we consider that the assimilation of $^{14}CO_2$ in the vegetation takes place after one month, two months, or three months. For a one-month delay, the Pearson correlation coefficient between the radiocarbon activity in air and vegetation was 0.14 with an R^2 value of 0.0189. For a two-month delay, the Pearson correlation coefficient was 0.23 and R^2 equal to 0.0543, while for a three-month delay, the Pearson correlation coefficient was −0.10 and R^2 equal to 0.0093.

Summary and conclusion

The presented study assesses the influence of a thermoelectrical power plant on environmental radiocarbon levels in the Govora Industrial Area. Due to the Suess effect, a relative decrease of the ^{14}C activity on a local scale was expected as a result of the dilution of the carbon isotopic mixture by fossil carbon. At the same time in the studied location, a nuclear facility designed for the detritiation of heavy water from CANDU reactors operates. Atmospheric ^{14}C is included in the environmental monitoring program due to the gaseous radioactive effluents of this installation.

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The sampling and preparation procedures presented have proven to be simple procedures both for $^{14}CO_2$ sampling into sodium hydroxide solution and for sample preparation by direct absorption method. The method involves capturing carbon dioxide from the sample in the scintillation cocktail and measuring it in an ultra-low level liquid scintillation spectrometer. LSC radiocarbon measurement is less expensive than other techniques but with the disadvantage of higher uncertainties compared with other techniques like accelerator mass spectrometry.

The radiocarbon results in the atmosphere had a decreasing trend, but due to local influence caused by the continuous production of fossil CO_2, we cannot observe a seasonal variation. The average value for the atmosphere was 0.226 ± 0.016 Bq/gC. ^{14}C values for vegetation were in the same range as those observed for the atmosphere. The minimums, maximums, and average of ^{14}C activity encountered in the air are found also in the vegetation.

A strong correlation between radiocarbon activity in air and vegetation was highlighted, with a Pearson correlation coefficient of 0.92. A good fit of the estimated trendline values to the measured values was highlighted also by an R^2 value of 0.8419.

Despite this good correlation, we considered three scenarios to see if the same correlation is maintained for a delay in the assimilation of $^{14}CO_2$ in the vegetation of one month, two months, or three months. The correlation between the activity of radiocarbon in the air and vegetation worsens for all these scenarios, which demonstrates that the radiocarbon in the atmosphere is immediately assimilated by the vegetation.

Author contribution. Faurescu Ionut and Varlam Carmen drafted the manuscript and did the interpretation of the experimental data. Faurescu Denisa, Vagner Irina, and Bogdan Diana performed the sampling and the radiocarbon measurements. Costinel Diana performed the $\delta^{13}C$ ratio measurements.

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