[RADIOCARBON, VOL 31, NO. 3, 1989, P 747–753]

^{14}C IN THE ENVIRONMENT OF SWISS NUCLEAR INSTALLATIONS

HEINZ HUGO LOOSLI and HANS OESCHGER

Physikalisches Institut der Universität, Sidlerstrasse 5
CH–3012 Bern, Switzerland

ABSTRACT. The results of a 10-year survey of ^{14}C content in tree leaves from the vicinity of Swiss nuclear installations are reported. The interpretation is based on the results from a reference station showing a systematic decrease from $\Delta^{14}C = +350‰$ in 1977 to ca +190 ‰ in 1987. Excess activities of up to ca 100‰ are observed in the vicinity of nuclear power plants which are compatible with release rates and dilution calculations. A higher excess of up to 1400‰ is measured in the close vicinity of a small research plant which releases the activity on top of the roof. The determined decrease of the activity with distance can be described by a power law with ca –1.4 in the exponent. Additional doses of ^{14}C to plants and population from excess ^{14}C activities are estimated to be negligible compared to natural doses.

INTRODUCTION

The ^{14}C content in leaves from beech trees have been monitored in the environment of Swiss nuclear installations since 1977 in collaboration with the Swiss Commission for the Survey of Radioactivity. Tree leaves are selected because they integrate the atmospheric activity over an extended period, mainly during their growth period in May and June. Short-term variations of atmospheric $^{14}CO_2$ concentrations are averaged out and long-term mean concentrations and effective additional dose commitments can be directly determined. The results can be compared with long-term dispersion model calculations for the effluents. The same type of tree (beech) is selected, as much as possible, to base the comparisons on the same growing season, *ie*, the same meteorological conditions.

Excess activities in the biosphere due to released $^{14}CO_2$ from nuclear installations are superimposed on the levels present at long distances from these emission points. However, these reference levels are influenced by various sources: the natural cosmic-ray-produced specific activity of 0.226 Bq/g of carbon is still elevated by ca 20% due to the remaining excess activity produced during the nuclear weapons testing mainly in the early 1960s. Also, a dilution of the natural ^{14}C level is caused by the release of ^{14}C-free CO_2 by burning of fossil fuels. Because of this Suess effect, the natural level is continuously decreasing to values ca 5–10% below the cosmic-ray-produced level. Annual variations are also observed close to big cities or industries (Levin, Münnich & Weiss, 1980; Levin *et al,* 1985, 1988). Thus, the ^{14}C results measured close to nuclear installations have to be compared with values at reference stations allegedly reflecting the same man-made influences except those from the investigated nuclear installations.

SUMMARY OF RESULTS

The measured $\Delta^{14}C$ results at reference stations are plotted in Figure 1. The principle station is ca 10km south of Bern, on the top of a hill, ca 400m

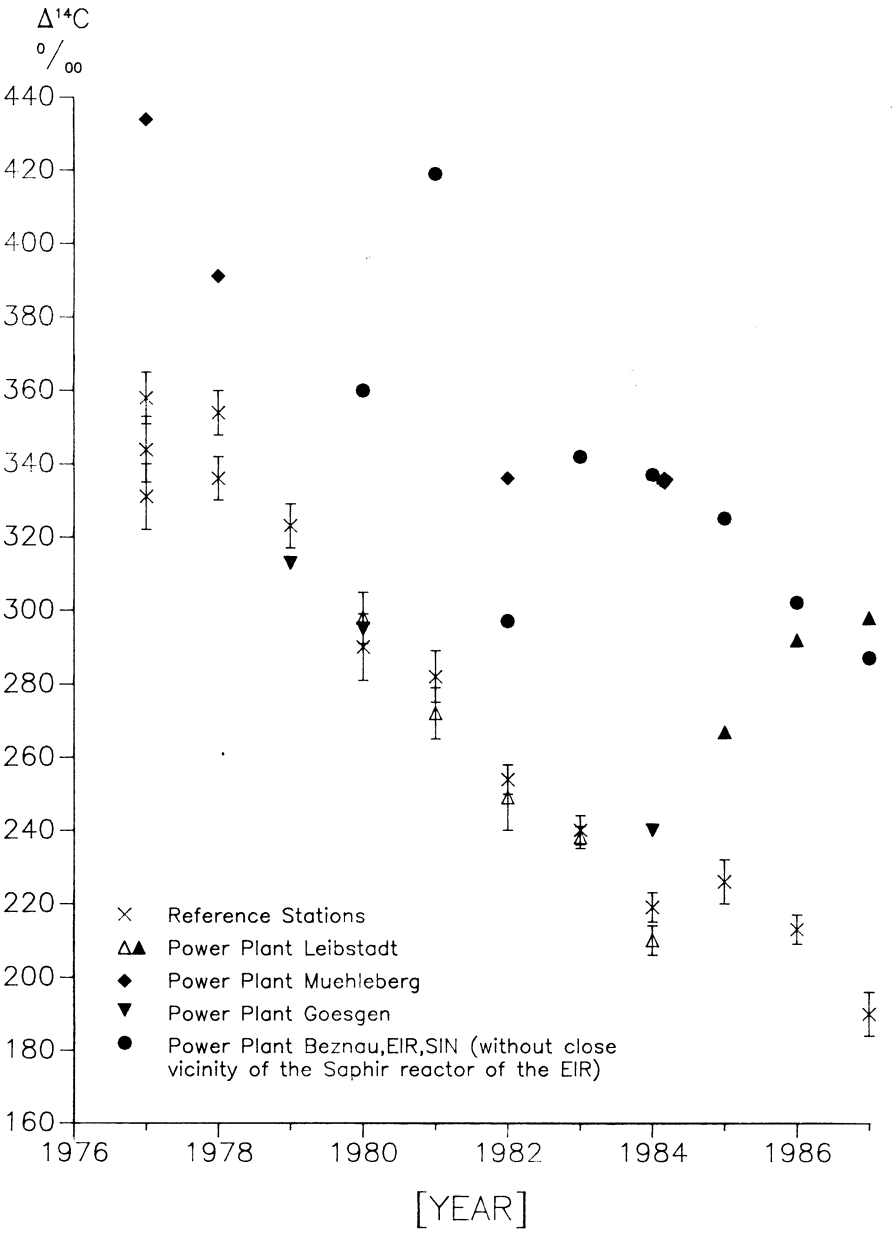


Fig 1. $\Delta^{14}C$ concentrations in beech-tree leaves at reference stations and maximum values of samples from the vicinity of Swiss nuclear power plants. Values at reference stations decrease because the ^{14}C activity produced by weapon tests is slowly disappearing into the oceans. The Gösgen PWR area shows less ^{14}C excess than the Leibstadt and Mühleberg BWRs. The maximum ^{14}C values measured near the two Beznau PWRs and the EIR and SIN nuclear installations are a combination of the release of these four installations.

higher than the city of Bern. From 1980 to 1984, a station near the Leibstadt power plant is also considered as a reference station, because this plant went into operation only in autumn 1984.

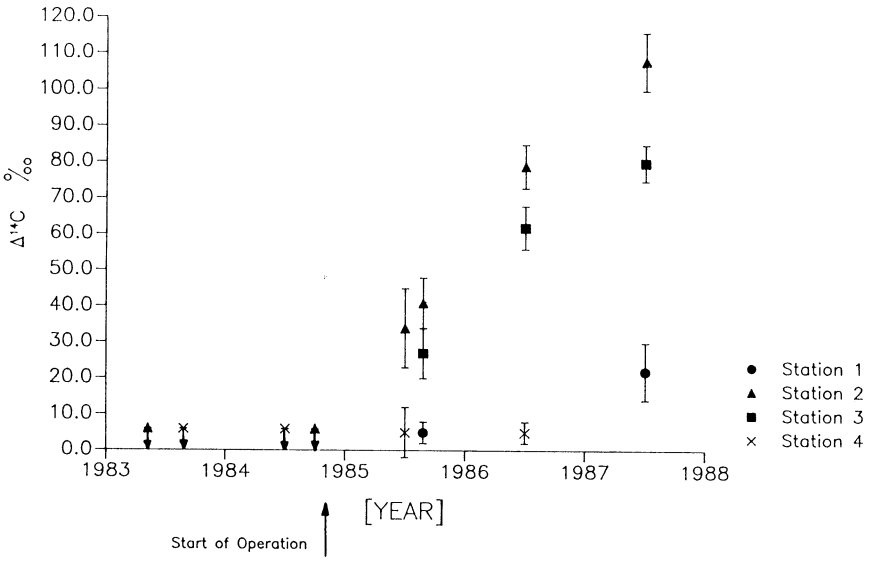
The $\Delta^{14}C$ values at reference stations are more or less linearly decreasing from 350‰ in 1977 to ca 190‰ in 1987, which indicates the speed at which excess bomb ^{14}C activity disappears from the atmosphere into the ocean. The measured values in tree leaves and the decrease rate agree with results obtained in CO_2 samples of clean atmospheric air (Levin *et al,* 1985, 1988). These results demonstrate that the values at our reference station are unaffected by local human activities.

Maximum concentrations in the area of nuclear installations which are plotted in Figure 1 are, in general, found between 0.5 and 1.5km in the main wind directions. The net values decrease rapidly with distance from these sites of maximum concentration. The excess ^{14}C in the vicinity of the Gösgen pressurized water reactor (PWR) is smaller than that of the Mühleberg and Leibstadt boiling water reactors (BWR) since BWRs emit a much larger fraction of ^{14}C in the form of CO_2 than PWRs. From 1977 to 1984, the excess values around Mühleberg remained quite constant at ca 60–120‰ above the base line of the reference stations. Emission rates, then, also remained constant within about a factor of 2.

Excess activities around two PWRs in Beznau, of the Eidgenössisches Institut für Reaktorforschung (EIR) and Schweizerisches Institut für Nuklearforschung (SIN) have to be attributed to releases from all four nuclear installations. The sites with maximum concentrations are centered among them. The measured net maximum values (excluding the very close surroundings of the Saphir research reactor) are 50–150 ‰ and again remained quite constant for the measuring period.

Excess ^{14}C activity near the Leibstadt power plant increased after it started operation in autumn 1984 (Fig 1, 2). The main wind directions (determined also by the Rhine valley) are to the northeast and southwest, which is also reflected in the spatial distribution of the measured $\Delta^{14}C$ values (Fig 2).

Although the estimated ^{14}C release rate from the Saphir research plant at EIR was established to be at least 5 times smaller than that from the two Beznau PWRs (KUeR, 1983), its immediate area was specially surveyed. This is because the activity is released at the roof, trees grow very close to the release point, and therefore, the dilution factor is expected to be much smaller than at the other sites. Tree leaf samples were collected at about emission height by the local fire brigade for samples 1 to 5 at distances from 30–150m (Fig 3). Excess ^{14}C values were quite high, especially in leaves of a tree located in the institute area. Results of two trees standing further apart but within the main wind directions are shown in Figure 3 (samples 6 and 7), where the sampling height was ca 2m above ground. Figure 3 shows that ^{14}C values remained constant within about a factor of 2 during the four years of observation. This range is probably also valid for the release rates.



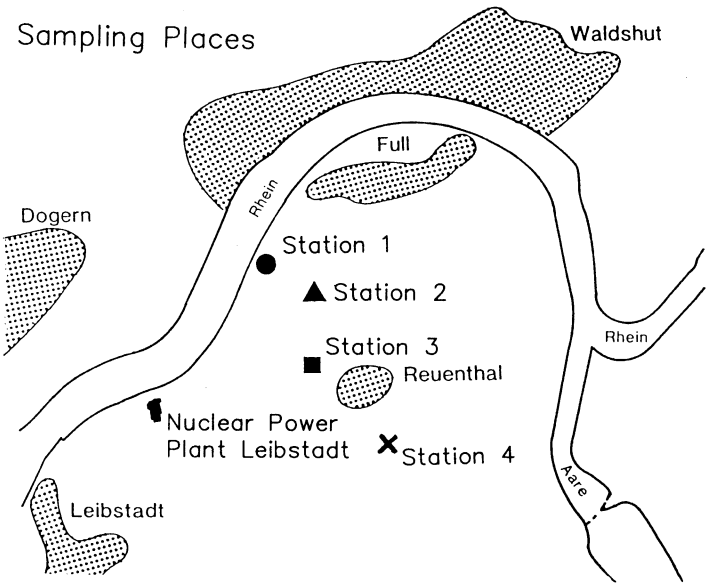


Fig 2. ^{14}C measurements in the vicinity of the Leibstadt nuclear power plant. Given are net values, relative to the reference station. Increased ^{14}C activity were measured in tree leaves after the power plant began operation in autumn 1984. The largest values are observed in the main wind direction, which is to the northeast of the plant. Increasing release rates are, at present, the most probable explanation for the increasing net activity from 1985 to 1987.

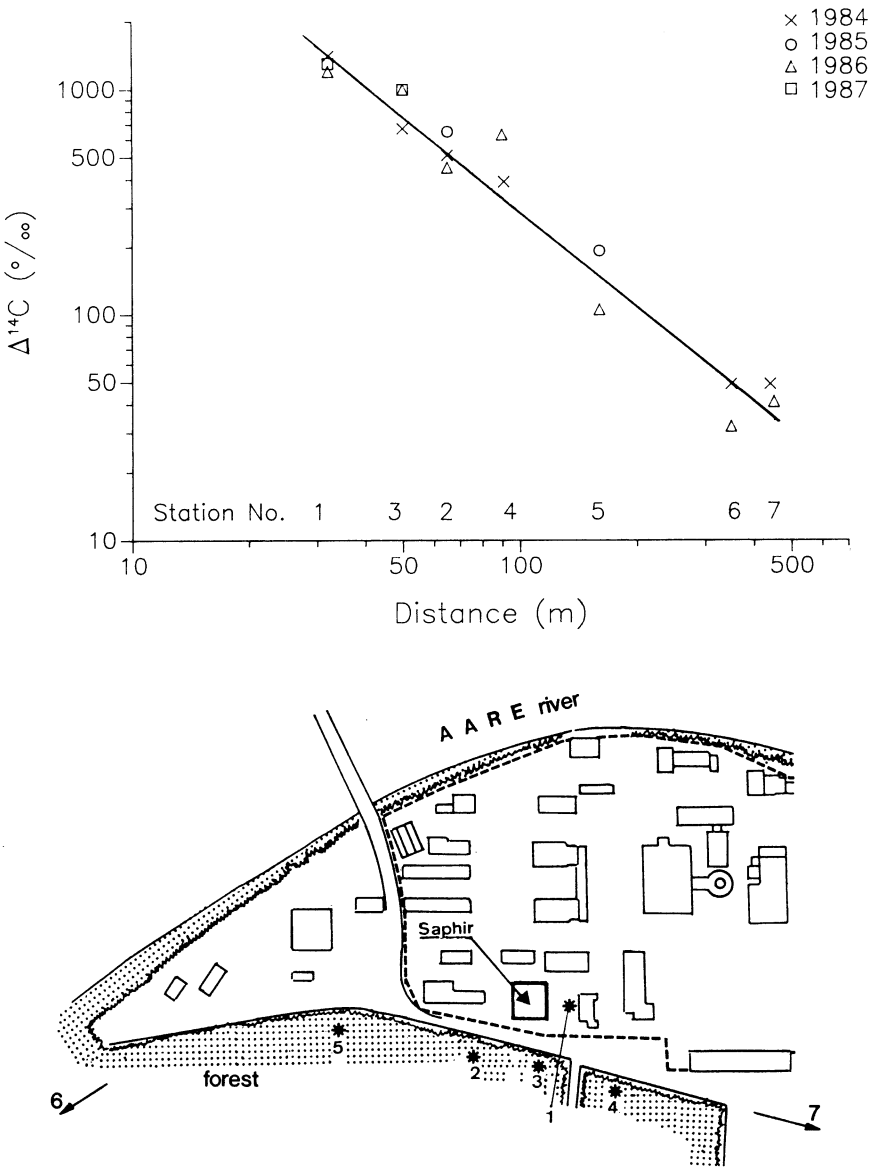


Fig 3. ^{14}C concentrations around the Saphir research reactor at EIR, given as net values relative to the reference station. Although the release rate is estimated to be below $7 \cdot 10^9$ Bq/yr, the measured excess values are quite high. This is because the ^{14}C activity is released at the roof and the sampling sites are very close to the emission point. The dilution factors are then smaller than at other sites. The samples were also collected at emission height. The decrease of the net activity with distance can be described by a power law.

DISCUSSION AND CONCLUSIONS

Measured excess ^{14}C activities in tree leaves can be compared with results of model calculations adopting release rates and dispersion coefficients based on stack height, weather statistics and topography. ^{14}C release rates by Swiss power plants and by the Saphir research reactor are not precisely known; the Swiss Nuclear Safety Authority, Hauptabteilung für die Sicherheit der Kernanlagen (HSK), estimates ca $4 \cdot 10^{10}$ Bq/y for Beznau, $1 \cdot 10^{11}$ for Gösgen, $4 \cdot 10^{11}$ for Leibstadt, $2 \cdot 10^{11}$ for Mühleberg and $<7 \cdot 10^{9}$ Bq/y for the Saphir reactor (KUeR, 1983). Less than 50% is released as CO_2 around the Beznau and Gösgen PWRs, which can be used for assimilation by plants.

Dispersion coefficients were determined for the vicinity of the Mühleberg power plant during the 1977 growing season yielding an agreement between measured and calculated ^{14}C values within a factor of 2 to 4 (Loosli *et al,* 1981; KUeR, 1978). For other plants in other areas, only annual average short- and long-term dispersion coefficients are known. Similar ^{14}C excess can be expected for both, Leibstadt and Mühleberg, as observed in 1987. The observed increase from 1985 to 1987 for Leibstadt, however, points to an increasing release rate, since it can be assumed that at least 80% of the organic material is newly formed by a leaf in the sampling year (Levin *et al,* 1988).

The dispersion of ^{14}C from the Saphir research reactor at EIR is a special case due to the short distance of the sampling sites from the release point. The measured concentrations can be described by a power law (Fig 3):

$$\Delta^{14}C(x) = \Delta^{14}C(d) \cdot \left(\frac{x}{d}\right)^{-1.4} = 1400‰ \cdot \left(\frac{x}{32\,m}\right)^{-1.4}$$

It is surprising that the results are on a straight line although the sampling sites are in various directions from the emission point and only generally in the main wind directions. The wind rose determined in the EIR area cannot be used for model calculations because it is measured on top of a large stack. More important is that the edge of the local forest may help in channeling the winds, especially in the direction of sampling points 6 and 7. More scatter of the excess ^{14}C values is observed at longer distances, which, however, may also reflect additional sources, especially from the Beznau PWRs (KUeR, 1987). The determined exponent in the power law of -1.4 is lower than theoretical values (eg, Turner, 1970). However, due to the special site and topography this may not be a real difference to the usually adopted values of -1.6 to -1.9 for average daytime weather categories and 0m emission height.

DOSES TO PLANTS AND POPULATION FROM EXCESS ^{14}C ACTIVITY

The maximum additional dose of ^{14}C in beech-tree leaves around the Saphir reactor due to its ^{14}C release is ca 0.01mGy per leaf lifetime. This is much smaller than the average natural doses of 0.5 – 5mGy for the same

period and also smaller than natural variations (Loosli & Weiss, 1988). No effects are therefore expected. No agricultural products are grown near the Saphir reactor.

If a farmer in the vicinity of a Swiss power plant would only eat vegetables, milk and meat with a ^{14}C concentration increased by 100 ‰ (compared with unaffected levels) its additional yearly effective dose equivalent would be in the order of 0.001 mSv. This is again negligible compared to natural dose variations and even more compared to natural doses: 1.3mSv/yr from cosmic ray, terrestrial and internal sources, and ca 2.2mSv from Radon daughters in Switzerland (KUeR, 1988).

ACKNOWLEDGMENTS

This work was supported by the Federal Commission for the Survey of Radioactivity and by the Swiss National Foundation. Thanks are due to Trudi Riesen and Markus Moell for sample preparation and careful measurements and to José Rodriguez and Ralf Weppernig for drawing the figures.

REFERENCES

KUeR, 1978, 1981, 1983, 1988, Annual reports of the Federal Commission for the Survey of Radioactivity to the Swiss Government: Fribourg, O Huber, ed.

Levin, I, Kromer, B, Barabas, M and Münnich, K O, 1988, Environmental distribution and long-term dispersion of reactor $^{14}CO_2$ around two German nuclear power plants: Health Physics, v 54, no. 2, p 149–156.

Levin, I, Kromer, B, Schoch-Fischer, H, Bruns, M, Münnich, M, Berdau, D, Vogel, J C and Münnich, K O, 1985, 25 years of trophospheric ^{14}C observations in central Europe: Radiocarbon, v 27, no. 1, 1–19.

Levin, I, Münnich, K O and Weiss, W, 1980, The effect of anthropogenic CO_2 and ^{14}C sources on the distribution of ^{14}C in the atmosphere, *in* Stuiver, M and Kra, R S, eds, Internatl ^{14}C conf, 10th, Proc: Radiocarbon, v 22, no. 2, p 379–391.

Loosli, H, Schriber, G, Moell, M, Oeschger, H and Riesen, T, 1987, C-14 activity measurements in tree leaves from the vicinity of Swiss nuclear power stations: Jahrestagung des Fachverbandes für Strahlenschutz, 14th, Lausanne, 30 Sept – 2 Oct 1981, Proc, p 348–355.

Loosli, H and Weiss, W, 1988, Weshalb kein ursächlicher Zusammenhang zwischen den Radioaktivitätsabgaben aus Kernkraftwerken und den Waldschäden besteht: Jahrestagung des Fachverbandes für Strahlenschutz, 19th, Salzburg, 15–19 Sept, Proc, p 248–262.

Turner, D B, 1970, Workbook of atmospheric dispersion estimates: Ed Environmental Protection Agency, no. AP-26, March 1972.