



RADIOCARBON AND ATMOSPHERIC ^{14}C PIONEER ATHOL RAFTER

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ABSTRACT. Direct atmospheric ^{14}C measurements began in New Zealand in 1954, initially to improve ^{14}C as a dating tool, but quickly evolving into a method for understanding the carbon cycle. These early ^{14}C measurements immediately demonstrated the existence of an “Atom Bomb Effect,” as well as an “Industrial Effect.” These two gigantic tracer experiments have been utilized via ^{14}C measurements over the years to produce a wealth of knowledge in multiple research fields including atmospheric carbon cycle research, oceanography, soil science, and aging of post-bomb materials.

KEYWORDS: atmosphere, bomb effect, Rafter, Suess effect.

INTRODUCTION

Athol Rafter and Gordon Fergusson started atmospheric ^{14}C measurements near Wellington, New Zealand, in late 1954 (Rafter 1955a). Initially, the goal was to improve ^{14}C as a dating tool by investigating the natural ^{14}C levels in the environment, including the atmosphere, surface ocean, and biosphere (Rafter 1955a). In a 1965 after-dinner speech at the Sixth International Radiocarbon and Tritium Conference in Pullman, Washington, Rafter recounted his introduction to the world of radiocarbon dating: “A few days later I was walking home quietly through the grounds of Parliament Buildings when coming in the opposite direction was the Head of our Department, a Mr Callaghan, who stopped me with the statement, ‘Rafter, I have just come from a meeting with geologists who tell me there is a method of dating by means of carbon that should be able to tell the age of our volcanic ash showers. Would you see if you could develop this method and stop the geologists arguing?’ I said a confused goodnight and continued on my way home somewhat more puzzled than usual” (Rafter 1965).

It wasn’t long before Rafter and compatriots Gordon Fergusson and George Page had an operational ^{14}C lab, making their first ^{14}C measurements in 1951 (Fergusson and Rafter 1953). These first measurements constrained the age of the Taupo supervolcano eruption “volcanic ash shower” and its ubiquitous tephra layer throughout New Zealand and the South Pacific, to about 170 AD. Unfortunately, these hard-won results did not stop the geologists arguing. The age was further constrained to 230 AD (Sparks et al. 1995), refined to 232 ± 5 AD (Hogg et al. 2011), and disputed again more recently (Holdaway et al. 2018; Hogg et al. 2019). Interestingly, when the 1951 measurements are calibrated, the result comes out remarkably close to the later results at 245 AD, albeit with a larger uncertainty (Sparks 2004).

Nonetheless, the arguing geologists no doubt inspired the early researchers in their efforts to achieve the highest quality and precision. Libby and his colleagues had achieved around 2% precision with their solid carbon measurement system (Anderson et al. 1947; Libby et al. 1949), but Rafter and Fergusson were stymied in their early attempts by the state of the local roads (Rafter 1953). Rafter would prepare solid samples at his laboratory in Wellington city, but the

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Table 1 The first three $^{14}\text{CO}_2$ measurements ever made, on samples collected as CO_2 absorbed into alkaline absorption at Makara, New Zealand. Results were originally reported as “enrichment in ^{14}C with respect to wood” (Rafter 1955a), and here are reported as $\Delta^{14}\text{C}$ according to Stuiver and Polach (1977), recalculated from the original counting data.

NZ	Collection period	$\Delta^{14}\text{C}$	$\Delta^{14}\text{C}$ unc.
2100	24 Nov–30 Dec 1954	−17.7	7.5
2099	31 Jan–16 Mar 1955	−10.1	7.6
2098	17 Mar–8 May 1955	−1.4	7.6

process of transporting the samples the 15 km or so to Fergusson’s counters in Lower Hutt (near the current location of the Rafter Radiocarbon Laboratory) would disappointingly result the transfer of the carbon carefully adhered to the walls of a copper cylinder to a pile of unusable rubble in the bottom of the container. Thus a better method was needed, and Rafter and Fergusson were the first to develop a CO_2 gas counting system, making their first measurements in 1951 (Rafter 1955b), although European researchers working on a similar system published their work earlier (see in this issue Grootes and van der Plicht 2021).

The CO_2 counting technique was not only easier and less dangerous, but the measurement precision was markedly improved over the earlier designs, achieving around 0.5% precision (de Vries and Barendsen 1953; Rafter 1955a) vs. the 2% obtained by Libby and others (Anderson et al. 1947). It was this improved precision that allowed detection of the beginning of the ^{14}C bomb spike from first three atmospheric ^{14}C measurements in 1954 and 1955 (Table 1, Figure 1). Yet “the CO_2 method [was] a most beautiful method and like all things beautiful has a most cantankerous side, the presence of electronegative impurities that drive technicians to despair, scientists to drink and harmony to discord” (Rafter 1965).

EARLY ATMOSPHERIC $^{14}\text{CO}_2$ RESULTS

The very first measurements at the New Zealand laboratory quickly led to a recognition that atmospheric $^{14}\text{CO}_2$ measurements would be invaluable in understanding the carbon cycle itself. Svante Arrhenius had worked out that fossil fuel emissions could double the amount of CO_2 in the atmosphere but expected it to take a few thousand years (Arrhenius 1896). Rafter’s early measurements (Rafter and Fergusson 1958), along with tree-ring reconstruction of 1940s and 1950s atmospheric $^{14}\text{CO}_2$ (Suess 1955), demonstrated a decrease in ^{14}C in the atmosphere that was hypothesized to be due to the absence of ^{14}C in the carbon from fossil fuel burning that had been added to the atmosphere (Revelle 1986), and that it was happening about 30 times faster than Arrhenius had expected (Suess 1955). Both Rafter and Suess recognized there was a need to measure atmospheric CO_2 mole fractions more directly, leading to the initiation of the iconic CO_2 mole fraction record at Mauna Loa, Hawai’i in 1958 (Keeling 1960). These discoveries by Arrhenius, Rafter, and Suess were the first two major science discoveries about climate change, and the fact that global changes can happen faster than expected in some early science papers has been rediscovered many times since.

Soon after, Rafter and Fergusson’s research led to another discovery, that the surface ocean appears about 400 years older than the overlying atmosphere (Rafter 1955a; Craig 1957). This key research set the stage for the marine radiocarbon calibration research field (Heaton et al.

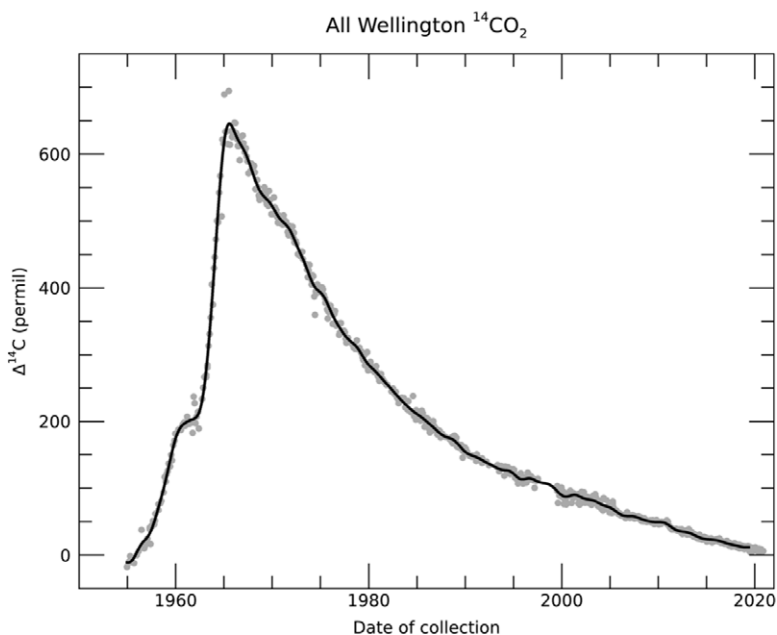


Figure 1 The Wellington $^{14}\text{CO}_2$ dataset, reported as $\Delta^{14}\text{C}$. Black line is a smooth curve fit to the measured data (gray points). The dataset has recently been updated, extending to 2021 and removing problematic measurements from the 1990s and 2000s (Rafter and Fergusson 1957b; Manning et al. 1990; Currie et al. 2011; Turnbull et al. 2017).

2020), and was the foundation for the use of ^{14}C in understanding the large-scale ocean circulation (Broecker et al. 1985).

Rafter was surprised by the first few atmospheric $^{14}\text{CO}_2$ measurement results, which showed a significant upward trend: “Whether this is due to an experimental error in the method of collection or to some other factor will be discussed when a greater number of samples have been studied” (Rafter 1955a). In fact, these were the first observations of the atmospheric $^{14}\text{CO}_2$ increase due to atmospheric nuclear weapons testing. Apparently Rafter already suspected the cause in 1955, in 1957 saying that “sampling was continued to test whether or not there was a seasonal variation or a possible ^{14}C enrichment of the atmosphere from atomic explosions” (Rafter and Fergusson 1957a). In an unpublished paper delivered at a conference in Dunedin, New Zealand in 1957, Rafter stated, “I would like to tell you the possibilities of two gigantic tracer experiments that we are witnessing in the world today and how if we are quick enough, we will be able to solve some interesting problems in mass movements of interest to oceanography and meteorology.” He foresaw the use of careful measurements of both the Suess and the bomb effects as valuable tools in geophysical research. An unverified report claims that during an international conference in the mid-1950s, Athol Rafter prudently withdrew a presentation that would have demonstrated the link with nuclear weapons testing after a late-night visit from government agents. Not daunted for long, Rafter and Fergusson published a paper entitled “The atom bomb effect” in the *New Zealand Journal of Science and Technology* in 1957, and then shared the results more widely in *Science* later that same year (Rafter and Fergusson 1957a; Rafter and Fergusson 1957b). Rafter claimed that should secret atmospheric nuclear

testing be carried out in the Pacific, his $^{14}\text{CO}_2$ measurements meant that New Zealand would know about it within days (Priestley 2012).

A 1965 letter from Hans Suess (at the University of California, San Diego) to Athol Rafter reads, “it is embarrassing for many of us that the rise in the carbon-14 concentration in the atmosphere due to artificial sources was first discovered and quantitatively measured by your laboratory for the southern hemisphere.” This dose of healthy competition was perhaps the spark for Northern Hemisphere researchers to begin atmospheric $^{14}\text{CO}_2$ observations later in the 1950s and early 1960s. Certainly, the measurements from both hemispheres have provided a treasure trove of data that continues to be applied across multiple disciplines.

THE LEGACY OF EARLY ATMOSPHERIC ^{14}C MEASUREMENTS

Once the first atmospheric ^{14}C measurements had demonstrated an increase in $^{14}\text{CO}_2$ content, attributed to atmospheric nuclear weapons testing, several laboratories became interested in this phenomenon and took up tropospheric $^{14}\text{CO}_2$ measurements at numerous locations around the world (Nydal and Lövseth 1983; Berger et al. 1987; Levin et al. 1985; Manning et al. 1990; Nydal and Gislefoss 1996) as well as in the stratosphere (Telegadas et al. 1972). More recently, detonation locations and magnitudes have been matched up with the $^{14}\text{CO}_2$ observations to estimate the total ^{14}C production from nuclear weapons (Hesshaimer and Levin 2000; Naegler and Levin 2006), a key parameter in establishing global radiocarbon budgets.

Once the threat of catastrophic ^{14}C production had passed, many atmospheric $^{14}\text{CO}_2$ measurement records were halted, likely due more to funding constraints than a lack of interest on the part of researchers. Rafter and his team continued the Wellington record (Manning et al. 1990), moving the sampling site from the original Makara location on New Zealand’s western coast near Wellington. The new site at Baring Head, on the south coast near Wellington, proved to be consistent with Makara for ^{14}C content, but a better choice for CO_2 mole fraction measurements, as it is much less influenced by the local biosphere (Lowe 1974; Lowe et al. 1979). The Wellington record continues to this day, its 67 years of measurements representing the longest direct atmospheric trace gas or isotope record anywhere in the world. In the 1990s, Rafter’s original gas counting method was phased out and replaced with AMS. The New Zealand laboratory contributed substantially to the development of AMS, particularly for atmospheric applications and development of the hydrogen graphitization technique (Lowe 1984; Lowe et al. 1988). The atmospheric record continued to use the sodium hydroxide absorption method of sample collection to ensure continuity of the record and these measurements have been supplemented with whole air flask samples since 2012. Fortuitously, Rafter’s successors including Graeme Lyon and Gordon Brailsford had collected and archived CO_2 from whole air flasks in the late 1980s and early 1990s, and these were able to replace anomalous outlier data for that period. Less fortunately, the early AMS measurements from 1995 to 2005 were made with incomplete correction for isotopic fractionation during sample preparation and measurement, and subsequent analysis has demonstrated that these data are biased high. Tree-ring measurements from Baring Head and nearby in the 2010s demonstrated that the early gas counting measurements from 1954 through to the late 1980s did not display any detectable biases or problems, a real testament to the careful efforts of the early researchers at a time when there were few options to validate or check their data (Turnbull et al. 2017).

The other notable uninterrupted time series of ¹⁴CO₂ are from the European Alps, initially at the Austrian high altitude site of Vermunt (Levin et al. 1985). Like the Wellington record, the sampling site was moved in the 1980s, in this case to Jungfraujoch in Switzerland, an even higher altitude site more favorable for CO₂ and other trace gas measurements (see in this issue Levin et al. 2021).

Together these Southern and Northern Hemisphere records have provided a strong constraint on interhemispheric mixing times (Kjellström et al. 2000; Land et al. 2002). These long records are key to constraining the global radiocarbon budget and elucidating carbon exchange processes (Randerson et al. 2002; Naegler and Levin 2006; Levin et al. 2010). They provide the backdrop for determining the rate of uptake of anthropogenic carbon into the oceans (Broecker et al. 1985; Hesshaimer et al. 1994; Caldeira et al. 1998; Key 2004; Peacock 2004; Sweeney et al. 2007), as well as in understanding the gas exchange rate with the oceans (Krakauer et al. 2006). A renewed interest in atmospheric ¹⁴CO₂ observations since the turn of the century has allowed further investigation into Southern Ocean carbon exchange, demonstrating that upwelled deep waters drive an atmospheric latitudinal gradient in ¹⁴CO₂ (Levin et al. 1987, 2010; Graven et al. 2012a, 2012b).

The atmospheric measurements are also leveraged in soil radiocarbon studies, for which the ¹⁴C bomb spike provides a vital control on turnover times (Trumbore 2000; Eglinton et al. 2021). Perhaps more familiar to the radiocarbon dating community, the direct atmospheric ¹⁴CO₂ records form the backbone of the “calibomb” dataset, used to calibrate post-bomb ¹⁴C measurements since 1950 (Hua et al. 2013, 2021). The measurements of the rapid changes in atmospheric ¹⁴CO₂ content since 1950 mean that samples can be aged quite precisely, often to within a single year. A wealth of applications include authentication of art objects; testing confiscated elephant ivory for violations of the CITES agreement (Uno et al. 2013; Cerling et al. 2016) and other wildlife forensic studies (Uno et al. 2013); authentication of bioplastic source (Telloli et al. 2019; Rogers et al. 2021); and even as a tool for determining the age of human remains for forensic investigations.

While Rafter and his compatriots were able to observe the changes in ¹⁴CO₂ in the clean Southern Hemisphere air near Wellington, they also noted decreases in ¹⁴CO₂ content. This was the imprint of ¹⁴C-free fossil fuel CO₂ introduced into the atmosphere, now coined the “Suess Effect” (Suess 1955). Several early papers documented the Suess Effect in pre-bomb tree-ring samples (Lerman et al. 1970; Tans et al. 1979; Stuiver and Braziunas 1998), but the enormous perturbation of the bomb spike masked the Suess Effect for much of the latter half of the 20th century. The renewed enthusiasm for atmospheric ¹⁴CO₂ measurements in the last 20 years is primarily focused on ¹⁴CO₂, widely recognized as the “gold standard” tracer for fossil fuel CO₂ emissions. The clean air measurements such as those at Wellington and Jungfraujoch provide a background constraint when determining the recently added fossil fuel CO₂ component. This application has become widespread, used to determine fossil fuel CO₂ emissions from individual point sources (Turnbull et al. 2016), cities (Djuricin et al. 2010; Ding et al. 2013; Newman et al. 2016; Niu et al. 2016; Turnbull et al. 2011, 2018), to nations and regions (Levin et al. 2003; Levin and Kromer 2004; Hsueh et al. 2007; Palstra et al. 2008; Riley et al. 2008; Van Der Laan et al. 2010; Cui et al. 2019; Basu et al. 2020; Lee et al. 2020).

This ongoing addition of ¹⁴C-free fossil-fuel CO₂ is predicted, if emissions continue apace, to result in an apparent atmospheric radiocarbon age of around 2500 years in 2100 (Graven 2015). While

not the most dire consequence of human perturbations to the atmosphere, it will certainly be an inconvenience to the radiocarbon dating community.

CONCLUSIONS

The sheer breadth of ongoing applications for the atmospheric ^{14}C measurements can be seen in the citation rate. The Wellington ^{14}C record started by Athol Rafter has been cited directly (Rafter and Fergusson 1957b; Manning et al. 1990; Currie et al. 2011; Turnbull et al. 2017) and indirectly through the calibomb dataset (Hua and Barbetti 2004; Hua et al. 2013) more than 1000 times. In recognition of his huge influence on the development of atmospheric ^{14}C and radiocarbon measurements in general, the Wellington laboratory was renamed the Rafter Radiocarbon Laboratory in 1993 in celebration of Athol Rafter's 80th birthday (Sparks 2004).

With the new technologies introduced in the last few years, and the prospect of in situ ^{14}C measurements on the horizon, atmospheric ^{14}C measurements can be expected to expand even further in the coming decades. Athol Rafter and his fellow researchers who began these measurements might be surprised at how widely used their ideas and data have become. But perhaps not, in Rafter's own words, "if these ^{14}C increases in the main reservoirs of the carbon cycle can be adequately studied in both hemispheres, it would be possible to evaluate exchange constants across the stratospheric-tropospheric, tropospheric-surface ocean water, surface ocean – deep ocean water, and interhemispheric differences" (Rafter and Fergusson 1957a).

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