

# Carbon-14 Background, Pathway, and Dose Calculation Analysis for Nuclear Power Plants: A Sourcebook for Accurate Carbon-14 Dose Calculations

2013 TECHNICAL REPORT



# Carbon-14 Background, Pathway, and Dose Calculation Analysis for Nuclear Power Plants

*A Sourcebook for Accurate Carbon-14 Dose  
Calculations*

All or a portion of the requirements of the EPRI Nuclear  
Quality Assurance Program apply to this product.

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## Product Description

Commercial nuclear power plants (NPPs) generate carbon-14 during operation, and release it in power plant effluents. This report explores the current state of carbon and carbon-14 science and understanding to identify and recommend any enhancements to carbon-14 dose calculation methodologies.

### **Background**

While the amount of carbon-14 released by NPPs is small compared to natural and other anthropogenic sources, it is important to accurately document it, along with the associated dose to the public. In the United States (U.S.), nuclear power plant operators use the guidance in Regulatory Guide 1.21, Revision 2 “Measuring, Evaluating, and Reporting Radioactive Material in Liquid and Gaseous Effluents and Solid Waste” (2009) and Regulatory Guide 1.109 Revision 1 “Calculation of Annual Doses to Man from Routine Releases of Reactor Effluents for the Purposes of Evaluating Compliance with 10 CFR Part 50, Appendix I” (1977). The International Atomic Energy Commission (IAEA) has published various guidance documents to support the international community in estimating dose to the public due to carbon-14, including IAEA Technical Reports Series No. 421, “Management of Waste Containing Tritium and Carbon-14” (2004), and IAEA Report 472 “Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments” (2010).

Global carbon and carbon-14 inventories and scientific understanding and data are dynamic. The amounts of carbon and carbon-14 in the atmosphere have changed, and continue to change over time due to human activities such as atomic bomb testing, use of fossil fuels for energy, etc. Such changes in global carbon and carbon-14 signatures and scientific knowledge about how carbon-14 incorporates in the human dose pathway impact the calculation of dose due to carbon-14 from NPPs. As such, carbon-14 dose calculation methodologies should be updated to reflect the latest science and understanding to ensure they provide accurate results.

### **Objectives**

To explore the current state of carbon and carbon-14 science and understanding to identify and recommend any enhancements to carbon-14 dose calculation methodologies.

## **Approach**

The project team conducted an extensive literature search to determine the current international state of knowledge regarding carbon-14 in the environment. The report provides a generic assessment of radiocarbon dating and accelerator mass spectrometry for the reader to understand carbon-14 measurements in the environment. Finally, the report discusses key findings of the research that impact or enhance the accuracy of carbon-14 dose calculation methodologies. In anticipation of any potential revision of Regulatory Guide 1.109, a comparison of the international state of knowledge with the contents of Regulatory Guide 1.109 was conducted to provide recommendations for enhancements to these carbon-14 dose calculation methods.

## **Results**

Some of the key variables for the calculation of dose due to carbon-14 in nuclear power plant effluents include the concentration of natural carbon dioxide in the atmosphere, the amount of vegetation mass that is natural carbon, local food consumption rates, and dose conversion factors. The amount of natural carbon dioxide has increased over time, and the use of current values for this parameter leads to accurate dose calculations. The amount of vegetation mass that is natural carbon varies with the type of plant. Some dose calculation methods allow the use of vegetation-specific values while others assign a single value. The use of vegetation-specific values that reflect the vegetation grown and eaten around the NPP supports accurate, site-specific dose calculations. Food consumption rates have changed and may vary internationally. A complete study on current food consumption rates in specific countries and local areas may inform more accurate carbon-14 dose calculations. Finally, the use of dose conversion factors based on the latest dose assessment science is important for accurate carbon-14 dose calculations.

## **Applications, Value, and Use**

Accurate calculation and reporting of doses to the public from nuclear power plant operations supports strong industry and stakeholder relationships, and also informs international research efforts. The findings of this research can be used by nuclear power plant operators to calculate more accurate, site-specific doses to the public due to carbon-14 from nuclear generating station effluents. These findings may also inform regulatory bodies when they revise applicable regulatory guidance.

## **Keywords**

Carbon-14	Environmental radioactivity
Gaseous effluents	Dose calculations
Atmospheric transport	Radiocarbon



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# Section 1: Introduction

## 1.1 Purpose and Scope

Commercial nuclear power plants generate carbon-14 during operation and release carbon-14 in power plant effluents. While the amount of carbon-14 released by nuclear power plants is small compared to natural and other anthropogenic sources of carbon-14, it is important to accurately document the amount of carbon-14 released from nuclear power plants and the associated dose to the public. Nuclear power plants calculate the carbon-14 dose to the public from effluents using methodologies developed and/or accepted by respective regulatory bodies.

The status of carbon-14 limits, monitoring, and reporting around the world varies. Nuclear power plants in various countries, such as Canada, have a history of measuring carbon-14 in effluents and reporting doses to the public. In the United States, even prior to 2010, some plant did estimate and report doses due to carbon-14 in their plant effluents. Their experiences and data are valuable to other countries developing their own methods. Other countries, such as Sweden and Korea, have conducted studies in measurement and estimation of carbon-14 from nuclear power plants. (EPRI, 2010; EPRI, 2011) The international community, especially in Europe, is also interested in carbon-14 emissions from nuclear waste disposal sites.

In the United States (US), nuclear power plant operators calculate and report the radiological doses resulting from carbon-14 ( $^{14}\text{C}$ ) in the nuclear power plant (NPP) effluents per revised guidance in Regulatory Guide 1.21 “Measuring, Evaluating, and Reporting Radioactive Material in Liquid and Gaseous Effluents and Solid Waste”. The United States Nuclear Regulatory Commission (US NRC) issued Revision 2 of Regulatory Guide 1.21 in 2009 that guides US NPP operators to evaluate whether carbon-14 is a principal radionuclide in gaseous effluents, and if so, to calculate and report doses associated to the carbon-14. (US NRC, 2009a) United States Nuclear Power Plants use Regulatory Guide 1.109 Revision 1 “Calculation of Annual Doses to Man from Routine Releases of Reactor Effluents for the Purposes of Evaluating Compliance with 10 CFR Part 50, Appendix I” (US NRC, 1977b) to calculate dose to the public due to carbon-14 in nuclear power plant effluents. The US NRC intends to update radiation protection related regulations and regulatory guides to reflect up to date dose assessment science and terminology. (US NRC, 2012) Regulatory Guide 1.109 may be revised as part of this process.

The International Atomic Energy Commission (IAEA) has published various guidance documents to support the international community in estimating doses to the public due to carbon-14. IAEA Technical Reports Series No. 421, “Management of Waste Containing Tritium and Carbon-14,” published in 2004, provides information for the management of radioactive waste and effluents containing  $^{14}\text{C}$ . This document provides technical information related to carbon-14 in the environment and generation from commercial nuclear power reactors. It also provides technical information on waste collection, separation, treatment, conditioning, and storage and/or disposal of tritium and carbon-14. (IAEA, 2004) IAEA Report 472 “Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments,” published in 2012, provides information for use in the dose assessment of radionuclides in routine commercial nuclear facility effluents. (IAEA, 2010) These IAEA documents are referenced throughout this current study as they provide recent scientific knowledge that impact carbon-14 dose calculations.

In 2009, the Electric Power Research Institute (EPRI) initiated research to provide technical guidance for the accurate reporting of carbon-14 and associated doses in nuclear power plant effluents. EPRI’s carbon-14 research addresses carbon-14 generation and release from nuclear power plants and the carbon-14 dose calculations. This current report represents a continuation of EPRI research documented in EPRI Report 1024827 “Carbon-14 Dose Calculation Methods at Nuclear Power Plants” which addressed the available regulatory guidance and industry practices used in calculating carbon-14 doses.

The purpose of this current project is to explore the current state of carbon and carbon-14 science and understanding to identify and recommend any enhancements to carbon-14 dose calculation methodologies. These findings can be used by nuclear power plant operators to calculate more accurate, site-specific doses to the public due to carbon-14 from nuclear generating station effluents. These findings may also inform regulatory bodies when they revise applicable regulatory guidance.

This research explores the current global carbon and carbon-14 inventories; current scientific understanding of carbon-14 incorporation into human dose pathways; and finally the calculation of carbon-14 dose due to power plant operations. An extensive literature search was conducted to determine the current international state of knowledge regarding  $^{14}\text{C}$  in the environment. A generic assessment of radiocarbon dating and accelerator mass spectrometry is provided for the reader to understand  $^{14}\text{C}$  measurements in the environment. Finally, the key findings of the research that impact or enhance the accuracy of carbon-14 dose calculation methodologies are discussed. In anticipation of any potential revision of Regulatory Guide 1.109, a comparison of the international state of knowledge with the contents of Regulatory Guide 1.109 is conducted to provide recommendations for enhancements in these  $^{14}\text{C}$  dose calculation methods.



## 1.2 Background

Carbon has two stable isotopes: carbon-12 and carbon-13, representing 98.93% and 1.07% of total carbon, respectively. Carbon-14 ( $^{14}\text{C}$ ) is a naturally occurring, radioactive isotope of carbon produced by cosmic radiation interactions in the upper atmosphere. The natural abundance of  $^{14}\text{C}$  is less than 0.1% of total carbon. (NCRP, 1985). The amount of  $^{14}\text{C}$  in the atmosphere increased significantly in the 1950s and 1960s due to nuclear weapons testing. Commercial nuclear reactors also produce  $^{14}\text{C}$ . The amounts of  $^{14}\text{C}$  that is produced naturally or were produced by nuclear weapons testing are much greater than those produced in commercial nuclear reactors. (US NRC, 2009a) The National Council on Radiation Protection (NCRP) estimated that the carbon-14 due to fall-out would continue to be about two orders of magnitude greater than that produced by commercial nuclear power plants for the foreseeable future (NCRP, 1985).

Carbon-14 is radiologically relevant due to its ease of incorporation into the global carbon cycle and its long half-life. The majority of anthropogenic  $^{14}\text{C}$  releases is gaseous and lead to increased levels of  $^{14}\text{C}$  in the terrestrial environment. This represents an important pathway for the incorporation of the radionuclide into both human and non-human populations (Magnusson et al., 2004).

Carbon-14 has a radioactive half-life of  $5730 \pm 40$  years.  $^{14}\text{C}$  decays to nitrogen-14 ( $^{14}\text{N}$ ) via  $\beta$  particle emission. The decay energy for this reaction is 156.476 keV. The mean energy of this  $\beta$  particle is 49.47 keV. (EPRI, 2010) Carbon-14 is produced by several primary neutron activation reactions (Davis, 1977) ( NCRP, 1985):

- $^{14}\text{N} (n, p) ^{14}\text{C}$ ,
- $^{13}\text{C} (n, \gamma) ^{14}\text{C}$ ,
- $^{13}\text{N} (n, p) ^{14}\text{C}$
- $^{15}\text{N} (n, d) ^{14}\text{C}$
- $^{17}\text{O} (n, ^4\text{He}) ^{14}\text{C}$  (Thermal Neutrons)
- $^{16}\text{O} (n, ^3\text{He}) ^{14}\text{C}$  (Fast Neutrons)

The  $^{14}\text{N} (n, p) ^{14}\text{C}$  reaction yields 99.632% of the total  $^{14}\text{C}$  produced in power reactors. Additional detail related to the specific reactions and production of  $^{14}\text{C}$  in nuclear reactors can be found in EPRI Report 1021106, “Estimation of Carbon-14 in Nuclear Power Plant Gaseous Effluents” (EPRI, 2010).

The most important of the above nuclear reactions for the production of  $^{14}\text{C}$  in the environment is also the  $^{14}\text{N} (n, p) ^{14}\text{C}$  reaction due to the interaction of cosmic radiation with the upper atmosphere. Upon formation in the upper atmosphere,  $^{14}\text{C}$  rapidly oxidizes to  $^{14}\text{CO}_2$ . The  $^{14}\text{CO}_2$  then becomes available for chemical reactions and photosynthesis within the biosphere. The result is  $^{14}\text{C}$  equilibrium between atmospheric  $^{14}\text{CO}_2$  and active chemical and biological systems within the biosphere.

The amount of environmental  $^{14}\text{C}$  produced by the  $^{14}\text{N} (n, p) ^{14}\text{C}$  reaction is not well known, and attempts at estimating the contributions from this reaction result in a wide variety of estimates. Many studies have also taken measurements in an attempt to quantify the production from this mechanism, but making accurate measurements is difficult, and concentrations will vary by location. (Roussel-Debet et al., 2006) (NCRP, 1985) (UNSCEAR, 1977) A recent production estimate for  $^{14}\text{C}$  from the above reaction from natural sources is  $1.54 \text{ PBq/yr}^1$  (Argonne National Laboratory, 2005). For additional discussion of  $^{14}\text{C}$  production and inventories in the environment refer to Section 4.

In Regulatory Guide 1.21 Revision 2, “Measuring, Evaluating, and Reporting Radioactive Material in Liquid and Gaseous Effluents and Solid Waste”, the United States Nuclear Regulatory Commission (NRC) staff provides estimates of the annual carbon-14 airborne releases from pressurized water reactors (PWRs) to be 8 to 9.5 curies per year per unit and boiling water reactors (BWRs) to be 5 to 7.3 curies per year per unit. (USNRC, 2009a) Detailed methods of estimating the  $^{14}\text{C}$  production rates in PWRs and BWRs are described in EPRI Report 1021106, “Estimation of Carbon-14 in Nuclear Power Plant Gaseous Effluents”<sup>2</sup>. (EPRI, 2010) Calculations using the EPRI method results in  $^{14}\text{C}$  production rates that vary from between 9 to 10 curies per unit per year (EPRI, 2012).

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<sup>1</sup> Note that the SI unit prefix P (or peta) indicates a multiplier of  $1 \times 10^{+15}$  and not to be confused with p (or pico) indicating a multiplier of  $1 \times 10^{-12}$ .

<sup>2</sup> The methodology developed by EPRI was reviewed and accepted by the Nuclear Regulatory Commission staff (EPRI, January 2011) as acceptable in meeting the guidance established in Regulatory Guide 1.21 (USNRC, 2009a) for the Annual Radioactive Effluent Release Reports.



## Section 2: United States Regulatory Bases for Carbon-14 Dose Calculation

The United States Nuclear Regulatory Commission (US NRC) intends to update radiation protection related regulations and regulatory guides to reflect up to date dose assessment science and terminology. (US NRC, 2012) These updates may impact carbon-14 dose assessment methodologies, for example, by the revision of Regulatory Guide 1.109 “Calculation of Annual Doses to Man from Routine Release of Reactor Effluents for the Purpose of Evaluating Compliance with 10 CFR Part 50, Appendix I,” which was previously revised in 1977. Since 1977, 35 year ago, there has been advances in scientific knowledge related to carbon and carbon-14 that, when implemented, will enhance the accuracy of carbon-14 dose calculations from nuclear power plant effluents. The results of this current EPRI research may inform any revisions of Regulatory Guide 1.109.

It should be noted, that even prior to revision of regulatory guidance, nuclear power plant operators may use alternative and/or site specific methods to comply with regulations and present these methods to the US NRC for review. The results of this current EPRI research will inform more accurate, site-specific carbon-14 dose calculations.

As such, a review United States carbon-14 regulatory history and regulatory guidance are provided in this section. This section also introduces some of the conservatisms associated with the current United States regulatory guidance with the objective of identifying opportunities for enhancing calculation accuracies. Applicable International Council on Radiation Protection (ICRP) recommendations are discussed.

### **2.1 Regulatory Guide 1.109 Guidance**

Regulatory Guide 1.109 Revision 1, “Calculation of Annual Doses to Man from Routine Release of Reactor Effluents for the Purpose of Evaluating Compliance with 10 CFR Part 50, Appendix I” (US NRC, 1977b) provides guidance for calculating doses to the public due to gaseous releases of carbon-14 from nuclear power plants. The detailed content of Regulatory Guide 1.109 and industry practices were discussed in detail in EPRI Report 1024827, “Carbon-14 Dose Calculation Methods at Nuclear Power Plants” (EPRI, 2012). The guidance in

Regulatory Guide 1.109 was available in 1977; however, due to the anticipated low contribution of carbon-14 to the overall dose from nuclear power plant effluents, the reporting of carbon-14 doses was not required at that time (Willis, 1981).

## **2.2 Regulatory Guide 1.21, Revision 2 (2009)**

In 2009, the U.S. NRC issued Revision 2 of Regulatory Guide 1.21 “Measuring, Evaluating, and Reporting Radioactive Material in Liquid and Gaseous Effluents and Solid Wastes.” In regulatory position 1.9 of this revised Regulatory Guide 1.21, the US NRC states that radioactive effluents have decreased since 1974 (when Revision 1 of Regulatory Guide was published) making it likely that carbon-14 is a principal radionuclide<sup>3</sup> in gaseous effluents. The US NRC guides nuclear power plant operators to evaluate whether <sup>14</sup>C is a principal radionuclide in their plant’s gaseous effluents and, if so, report carbon-14 dose in annual effluent reports. (US NRC, 2009a) Starting in 2010, nuclear power plants in the United States report higher doses to the public in their Annual Radiological Effluent Reports due to the carbon-14 being included in dose estimation calculations even though there were no actual unexpected or abnormal increases in carbon-14 in effluents<sup>4</sup> (EPRI, 2012; US NRC, 2013).

Regulatory Guide 1.21, Revision 2 (US NRC, 2009a), in regulatory position 5.9, references the following Regulatory Guides as being acceptable models that should be used in dose calculations:

- 1.109 “Calculation of Annual Doses to Man from Routine Release of Reactor Effluents for the Purpose of Evaluating Compliance with 10 CFR Part 50, Appendix I” (US NRC, 1977b)
- 1.111 “Methods for Estimating Atmospheric Transport and Dispersion of Gaseous Effluents in Routine Releases from Light-Water-Cooled Reactors” (US NRC, 1977a)
- 1.112 “Calculation of Releases of Radioactive Materials in Gaseous and Liquid Effluents from Light-Water-Cooled Power Reactors” (US NRC, 2007)

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<sup>3</sup> Regulatory Guide 1.21 Revision 2 (USNRC, 2009a) introduces the term “principal radionuclide” in a risk-informed context. A licensee may evaluate the list of principal radionuclides for use at a particular site. The principal radionuclides may be determined based on their relative contribution to (1) the public dose compared to the 10 CFR 50 Appendix design objectives or (2) the amount of activity discharged compared to other site radionuclides. If adopting a risk-informed perspective, a radionuclide is considered a principal radionuclide if it contributes either (1) greater than 1 percent of the 10 CFR Part 50, Appendix I, design objective dose for all radionuclides in the type of effluent being considered, or (2) greater than 1 percent of the activity of all radionuclides in the type of effluent being considered. Regulatory Guide 1.109 lists the three types of effluent as (1) liquid effluents, (2) noble gases released to the atmosphere, and (3) all other radionuclides released to the atmosphere.

<sup>4</sup> Refer to EPRI Report 1024827 Dose Calculation Methods at Nuclear Power Plants (EPRI, 2012) for additional details. Also refer to the NRC web pages for the details for each nuclear generating station Annual Radiological Effluent Report (<http://www.nrc.gov/reactors/operating/ops-experience/tritium/plant-info.html>) (USNRC, 2013).

- 1.113 “Estimating Aquatic Dispersion of Effluents from Accidental and Routine Reactor Releases for the Purpose of Implementing Appendix I” (US NRC, 1977d)

This guidance establishes the dose calculation methods used by most all United States licensees.

Regulatory Guide 1.21, Revision 2 also states, “The quantity of C-14 discharged can be estimated by sample measurements or by use of a normalized C-14 source term and scaling factors based on power generation.” The guidance identifies NCRP Report 81 (NCRP, 1985) and NUREG-017 “Calculation of Releases of Radioactive Materials in Gaseous and Liquid Effluents from Pressurized Water Reactors PWR-GALE Code” (US NRC, 1985b) as acceptable methods. Based on a public meeting held on January 20, 2011 (EPRI, 2011a), the commission staff agreed to also accept the method developed by the Electric Power Research Institute in “Estimation of Carbon-14 in Nuclear Power Plant Effluents” (EPRI, 2010).

For assessment of doses to the public due to nuclear power plant effluents (including carbon-14), Regulatory Guide 1.21 states:

If bounding assessments are not used, licensees should perform evaluations to determine the dose to a real, maximum exposed member of the public. A member of the public is typically a real individual in a designated location where there is a real exposure pathway (e.g., a real garden, real cow, real goat, or actual drinking water supply) and is typically not a fictitious fencepost resident or an exposure pathway that includes a virtual goat or cow. Licensees are encouraged (but not required) to use real individual members of the public when performing dose assessments for radioactive discharges (US NRC, 2009b).

It further states that “real exposure pathways are identified for routine discharges and direct radiation based on the results of the land use census”. Further guidance regarding the performance of a land use census can be found in NUREG-1301 “Offsite Dose Calculation Manual Guidance: Standard Radiological Effluent Controls for Pressurized Water Reactors” or NUREG-1302 “Offsite Dose Calculation Manual Guidance: Standard Radiological Effluent Controls for Boiling Water Reactors” (US NRC, 1991a, 1991b).

## **2.3 Maximum Exposed Individual**

Dose calculations require estimates or data related to incorporation of radionuclides (in this case carbon-14) into environmental media and the uptake of the radionuclides by an individual through the environmental media. Useful information regarding the uptake by the age groups defined in Regulatory Guide 1.109 is discussed in Section 5 of this report. Section 4 of this report also discusses transfer coefficients used in compartment dose models. There exist conservatism in the methods of Regulatory Guide 1.109, most notably the

introduction of the term maximum exposed individual. This is “characterized as maximum with regard to food consumption, occupancy, and other usage of the region in the vicinity of the plant site and as such represent individuals with habits representing reasonable deviations from the average for the population in general.” Regulatory Guide 1.109 further states “that where site-specific information and data is used, its justification should be documented for the NRC staff’s review”. This guidance by NRC has introduced some flexibility in Regulatory Guide 1.109 that the licensee “may take into account any real phenomena or actual exposure conditions” (EPRI, 2012).

## **2.4 Evolving Science for Dose Factors**

Regulatory Guide 1.109 currently uses Internal Dose Commitment Factors (DCF or dose factors) that are based on International Council on Radiation Protection (ICRP) Publication 2 “Permissible Dose for Internal Radiation” (ICRP, 1959). More modern dose factors are provided in ICRP Publication 72 “Age-dependent Doses to the Members of the Public from Intake of Radionuclides - Part 5 Compilation of Ingestion and Inhalation Coefficients” (ICRP, 1996) and have been adopted as the international standard for calculations of dose to the member of the public. A robust comparison of the methods and differences in the dose factors presented by ICRP 2 and ICRP 72 can be found in Appendix A to EPRI Report 1024827, “Carbon-14 Dose Calculation Methods at Nuclear Power Plants” (EPRI, 2012). In summary this report states, the more modern dose factors from ICRP 72 are based on updated and modern science and research and yield dose estimates that are considerably less than those calculated using ICRP 2 dose factors from Regulatory Guide 1.109. The following points emphasize the benefits of upgrading from the outdated ICRP 2 approach:

- The ICRP 72 model yields dose factors that are comparable across all organs within a given age class, implying that there is no non-stochastic “critical organ” exposure as would be indicated by the heavily-weighted ICRP 2 dose factors for bone.
- While any doses calculated with ICRP 72 DCFs result in doses that clearly fit into assumptions for stochastic exposure, the ICRP 2 DCFs would result in bone doses three to five times higher than those for the other organs, suggesting ICRP 2 more closely mimics a non-stochastic approach.
- The ICRP 72 approach is based on metabolic models developed in the 1990s, and represent science that is nearly four decades newer than the bases behind ICRP 2.
- When coupled with the NRC's adoption of establishing effluent concentrations limits based on stochastic dose effects because “... *non-stochastic effects are presumed not to occur at the dose levels established for individual members of the public...*” (10CFR20, Appendix B), it would appear that application of ICRP 72 dose factors in calculating carbon-14 doses is more technically-correct when calculating dose to a member of the public.

Such an adoption of ICRP 72 dose factors or proposed revisions to NRC regulations based on ICRP 103 “2007 Recommendations of the International Commission on Radiological Protection” (ICRP, 2007) would more accurately represent the resulting dose used to demonstrate compliance with 10 Code of Federal Regulations (CFR) Part 20 “Standards for Protection Against Radiation” and also with 10 CFR Part 50 “Domestic Licensing of Production and Utilization Facilities.” This would also align the dose assessment methodology used in the United States to that of the international radiation protection community.

This EPRI study reviews some of the technology behind the US NRC guidance contained in Regulatory Guide 1.109 with regard to carbon-14 dose calculations and provides current references and information that offer technical enhancements. The current guidance is now more than 35 years old and may be revised in a normal review cycle at NRC or as part of the proposed implementation of ICRP 103 (ICRP, 2007).







## Section 3: Radiocarbon Science and Laboratories

Background on radiocarbon science and laboratories is important in understanding carbon-14 ( $^{14}\text{C}$ ) in the environment since much of the information about the detailed behavior of  $^{14}\text{C}$  comes from the research, literature, and publications from these laboratories. Section 4 Radiocarbon in the Environment and Environmental Models in this report is supported by presenting the methods and units used by radiocarbon science. An excellent history of radiocarbon dating, standards, and equipment can be found in The Remarkable Metrological History of Radiocarbon Dating [II] (National Institute of Standards and Technology, 2004).

### 3.1 History of Radiocarbon Dating and Radiocarbon Units

In the 1950s Willard Libby and others at the University of Chicago successfully demonstrated the utility of using  $^{14}\text{C}$  decay to estimate the amount of time past since naturally occurring organic materials that were once part of a living system. This gave birth to the industry of radiocarbon dating which today is a primary chronometer for archeologists and geologists studying events within the last 50,000 years (Anderson and Libby, 1951).

Anderson and Libby (1951) originally assumed that the specific activity of  $^{14}\text{C}$  was a constant. It is well documented, however, that atmospheric production rates vary with variations in earth's cosmic flux influenced by solar winds and the earth's geomagnetic field. Long term variations are detectable in tree rings and ancient corals (Key et. al., 2001).

With the publication of U.S. Nuclear Regulatory Commission Regulatory Guide 1.21, Revision 2 "Measuring, Evaluating, and Reporting Radioactive Material in Liquid and Gaseous Effluents and Solid Waste" (USNRC, 2009a) and Regulatory Guide 4.1, Revision 2 "Radiological Environmental Monitoring for Nuclear Power Plants" (USNRC, 2009b) the interest in the better assessment of the environmental uptake of  $^{14}\text{C}$  in effluent of nuclear power plants has increased. The use of radiocarbon dating laboratories for this application is not new. It has been applied both academically and industrially for over four decades. As such, existing expertise is readily available for both sample collection strategies and the understanding of analytical error constraints are well established.

Radiocarbon ( $^{14}\text{C}$ ) is present naturally in a relative abundance of only  $1 \times 10^{-12}$  of total carbon. Present day activity in the atmosphere is only about 0.237 Bq/g of carbon (including natural sources and roughly a 5% present day elevation primarily due to thermonuclear weapons testing) (Mook and van der Plicht, 1999). Measurement of  $^{14}\text{C}$  at such low levels benefits from the specialties of a radiocarbon dating laboratory that only analyses natural levels of  $^{14}\text{C}$ . Conventional radiological laboratories used for radiological effluent measurements are not suited for this application due to their exposure to high background and highly elevated  $^{14}\text{C}$  in the laboratory. Conversely, a radiocarbon dating laboratory can never accept direct submission of radiological effluent samples.

Understanding how radiocarbon measurements serves to quantify  $^{14}\text{C}$  for environmental uptake of nuclear generating unit  $^{14}\text{C}$  effluent rests in understanding how radiocarbon dating works. This Section limits discussion to providing clarity on the use of results and units of radiocarbon reporting. Since this application has been in use for 70 years with hundreds of articles and many books on the subject, ample resources are available to fully understand the process. A very good summary and review on radiocarbon dating can be found in “Radiocarbon: A Chronological Tool for the Recent Past” (Hua, 2009).

Understanding the units provided by radiocarbon dating laboratories is of great importance. Being a mature and historically non-standardized industry which is largely comprised of teaching laboratories, reporting and use of results in conflicting units has characterized the field for decades. This section of the report will give guidance on how to use ambient  $^{14}\text{C}$  results as they relate to radiological environmental measurements for nuclear utility usage, we forego some of the history related this. For those interested, a very good summary and review of units is provided in the reference list (Stenström, 2011). This document focuses discussion on units of “fraction of modern” ( $F^{14}\text{C}$ ) and “percent modern carbon” (pMC). These are the units from which specific activity calculations can be readily made.

The conventions of radiocarbon dating relate the  $^{14}\text{C}$ /stable C ratio of the material relative to a “modern” reference standard followed by application of the age equation. See Equation 3-1 below:

$$\text{Radiocarbon Age} = -8033 * \ln \left( \frac{\frac{^{14}\text{C}_{\text{sample}}}{^{12}\text{C}_{\text{sample}}}}{\frac{^{14}\text{C}_{\text{modern reference}}}{^{12}\text{C}_{\text{modern reference}}}} \right) \quad \text{Eq. 3-1}$$

Where:

8033 = the inverse of the decay rate ( $1/\lambda$ ) associated with the Libby half-life of 5568 years<sup>5</sup>.

sC = stable carbon ( $^{12}\text{C} + ^{13}\text{C}$ )

Modern reference refers to NIST<sup>6</sup>-4990b, NIST3-4990c or IAEA<sup>7</sup>-C6 radiocarbon dating standards. Detailed descriptions and history of these standards can be found in The Remarkable Metrological History of Radiocarbon Dating [II] (National Institute of Standards and Technology, 2004).

Only the ratio in Equation 3-1, between the sample and the reference, is of significance for the purposes of specific activity calculations. This ratio is linear and is termed “Fraction of Modern” ( $F^{14}\text{C}$ ). It is often reported as a percentage by multiplying  $F^{14}\text{C}$  by 100, termed percent modern carbon (pMC).

$$\text{Fraction of Modern Carbon } (F^{14}\text{C}) = \left( \frac{\frac{{}^{14}\text{C}_{\text{sample}}}{s\text{C}_{\text{sample}}}}{\frac{{}^{14}\text{C}_{\text{modern reference}}}{s\text{C}_{\text{modern reference}}}} \right)$$

Eq. 3-2

$$\text{Percent Modern Carbon (pMC)} = F^{14}\text{C} * 100$$

Eq. 3-3

The absolute values assigned to the modern reference standards (above) are taken as of AD 1950. Frequently in papers associated with environmental studies the final result will take into account the decay in the reference standard since AD 1950. Convention defines the reporting unit “ $\Delta^{14}\text{C}$ ” to represent this decay-corrected result, in parts per thousand (aka “per mil”, o/oo). Per mil results are also frequently reported as  $\Delta^{14}\text{C}$ . In this case, per mil results are reported without reference decay taken into account (It is these two units that have most frequently been incorrectly reported in the literature). Specific activity as a fraction to total carbon can be calculated directly from fraction modern ( $F^{14}\text{C}$ ). And since pMC is simply  $F^{14}\text{C} \times 100$ , these two values ( $F^{14}\text{C}$  and pMC) are the values of interest in calculating specific activity.

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<sup>5</sup> The half-life of c-14 is  $5730 \pm 40$  years (Nature, 1962) which was adopted at the Fifth Radiocarbon Dating Conference held in Cambridge, England in 1962. The standard reference for radiocarbon dates at the time prior to Libby was the Journal Radiocarbon published by Yale University. Adoption and continued use of the Libby half-life was made at the Eight Radiocarbon Dating Conference in Wellington, NZ in 1972 and still stands today (Radiocarbon, 1977).

<sup>6</sup> National Institute of Standards and Technology.

<sup>7</sup> International Atomic Energy Agency.

### **3.1.1 Conversion of Percent Modern Carbon to Specific Activity**

When converting the pMC result to an absolute value such as Bq/Kg C (Becquerel per kilogram carbon) or pCi/Kg C (picocuries per kilogram carbon), the specific activity of the primary modern reference (HOxI, see Section 3.2 regarding this term) is used. This is taken as 226 +/- 1 Bq/Kg C, equivalent to 95% the activity of the specific batch Oxalic Acid number 1 measured in AD 1950.

Calculation of the specific activity for a sample is made by adjusting the sample pMC for <sup>14</sup>C decay since AD 1950 and multiplying that value by 226 Bq/kg C. This may be done according to the following formulas.

IMPORTANT: The following formulas provide specific activity per mass of CARBON.

#### ***Conversion of pMC to Bq/Kg C***

$$A = F^{14}C * e^{\frac{1950-y}{8267}} * 226 \quad \text{Eq. 3-4}$$

Where:

A = Specific Activity in Bq/Kg C

F<sup>14</sup>C = pMC/100

e = 2.71828

226 Bq/kg C

y = year of harvest/measurement

#### ***Conversion of pMC to pCi/Kg C***

$$A = F^{14}C * e^{\frac{1950-y}{8267}} * 6108 \quad \text{Eq. 3-5}$$

Where:

A = Specific Activity in pCi/Kg C

F<sup>14</sup>C = pMC/100

e = 2.71828

y = year of harvest/measurement

6108 pCi/Kg C = 226 Bq/Kg C x 27.027 pCi/Kg C/1 Bq/Kg C

## 3.2 Radiocarbon Analysis and Standards

Since the natural abundance of  $^{14}\text{C}$  is extremely low ( $1 \times 10^{-10} \%$  C), specialized analytical methods are required for environmental samples. These methods include synthesis of carbon to benzene ( $\text{C}_6\text{H}_6$ ) with subsequent counting in a liquid scintillation counter (LSC) or by direct counting in a particle accelerator (AMS). Historically gas proportional counting of  $\text{CO}_2$  has also been viable. Since detailed discussions of these methods can be found in both ASTM-D6866 “12 Standard Test Methods for Determining the Biobased Content of Solid, Liquid, and Gaseous Samples Using Radiocarbon Analysis” (ASTM, 2012), and CEN 15440 “Solid Recovered Fuels- Methods for the Determination of Biomass Content” (Comite’ Europeen de Normalisation, 2011), the discussion here is limited to the basic information required to understand results as they apply to a nuclear power plant radiological environmental monitoring program (REMP).

The natural abundance of stable carbon  $^{12}\text{C}$  is 98.8 % and  $^{13}\text{C}$  is 1.2 %. Depending upon the method of analysis, the analytical measure will be as  $^{14}\text{C}/^{13}\text{C}+^{12}\text{C}$  (LSC) or the  $^{14}\text{C}/^{12}\text{C}$  or  $^{14}\text{C}/^{13}\text{C}$  (AMS). The  $\text{F}^{14}\text{C}$  or pMC value obtained will be the same regardless of which ratio it was derived from.

The primary “modern” reference standard, manufactured at the time of Libby is oxalic acid derived from sugar beets (originally NBS-4990b now NIST-4990b, also known as Oxalic Acid I, OxI, and HOxI). It was determined that a factor of 0.95 times the measured  $^{14}\text{C}$ /stable carbon for HOxI approximated the year 1950. As such, 1950 was termed “modern”, and a value of 0.95 the measured value of the HOxI is termed 100 percent modern carbon (pMC).

The measure in the laboratory is purely relative. First a value for the reference standard is measured, and then a value for the sample is measured. The ratio between the two is then calculated for  $\text{F}^{14}\text{C}$ , and multiplied by 100 for pMC. Since measurement is being made on materials of biological origin, this value will include normalization of the carbon isotopes so that isotopic effects associated with different metabolic pathways are factored out. These effects are termed “isotopic fractionation” and the variable associated with the measure is termed “delta  $^{13}\text{C}$ ” ( $\Delta^{13}\text{C}$ ).

### 3.2.1 Accuracy and Precision

By convention, precision on the pMC values reported to the client by a laboratory will be one relative standard deviation. For analysis by Accelerator Mass Spectrometry (AMS) this should typically be  $< 0.5$  pMC for values less than 100 pMC, 0.5 – 2.0 for values between 100 – 200 pMC and  $> 2.0$  for values greater than 200 pMC. Be aware that precision quotes larger than these may indicate significant determinant error in the measurement.

The error cited by the laboratory for the pMC value is strictly counting error and by convention is reported as 1 sigma relative standard deviation. An established ambient level C-14 laboratory should have a *total laboratory error* below 2 sigma counting reproducibility. As such, two results can be considered statistically the same if they are within two sigma of each other.

It is important to remember the pure relative nature of the pMC result. No absolutes are being determined in the laboratory other than the weight of benzene for LSC measurements. The  $^{14}\text{C}$ /stable carbon ratio is measured for the NIST standard, the  $^{14}\text{C}$ /stable carbon ratio is measured for the sample, and a ratio is taken between the two for the pMC result (factoring out background signals and isotopic fractionation). Accuracy depends upon the underlying condition that the measurements on each were obtained under systematically the same conditions of chemistry and counting.

AMS radiocarbon measurements are made on graphite synthesized from carbon within the sample. The process involves first drying the sample, then sub-sampling a portion for conversion to  $\text{CO}_2$ , then synthesis of graphite as the medium for measure in the AMS. This process is identically performed on the modern reference material and a radiocarbon-dead material (serving for background subtraction). The accuracy of the results is highly dependent upon (1) the chemical background to be identical for all three, (2) the consistency of the graphite to be identical for all three, and (3) measurement of all three are made within the particle accelerator under identical calibration conditions.

Figure 3-1 illustrates the capability of AMS radiocarbon dating to systematically reproduce results to within 2 Bq/Kg C for a working standard of 121 Bq/Kg C. Figure 3-2 illustrates the capability of AMS radiocarbon dating to systematically measure accurate results on a known value reference material of  $128 \pm 1$  Bq/Kg C. Such capability can be expected for real-life environmental samples when modern reference, sample and background measurements are made on graphite produced under identical chemical conditions and measured quasi-simultaneously within the AMS. Figure 3-1 is a demonstration of reproducibility from a radiocarbon dating laboratory. 224 measurements on an internal working standard reproducing within  $\pm 2$  Bq/KgC are shown. Figure 3-2 is a demonstration of the accuracy from a radiocarbon dating laboratory. 78 measurements on an international radiocarbon standard of known  $^{14}\text{C}$  content (FIRI-I, (2)) are shown.

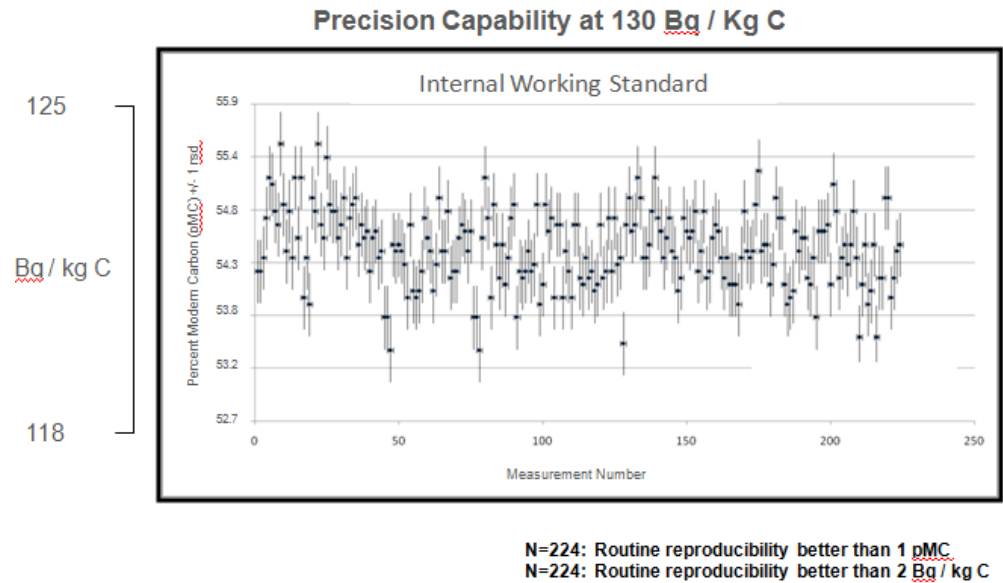


Figure 3-1  
Example of AMS Precision Capability (Provided by Beta Analytic, Inc.)

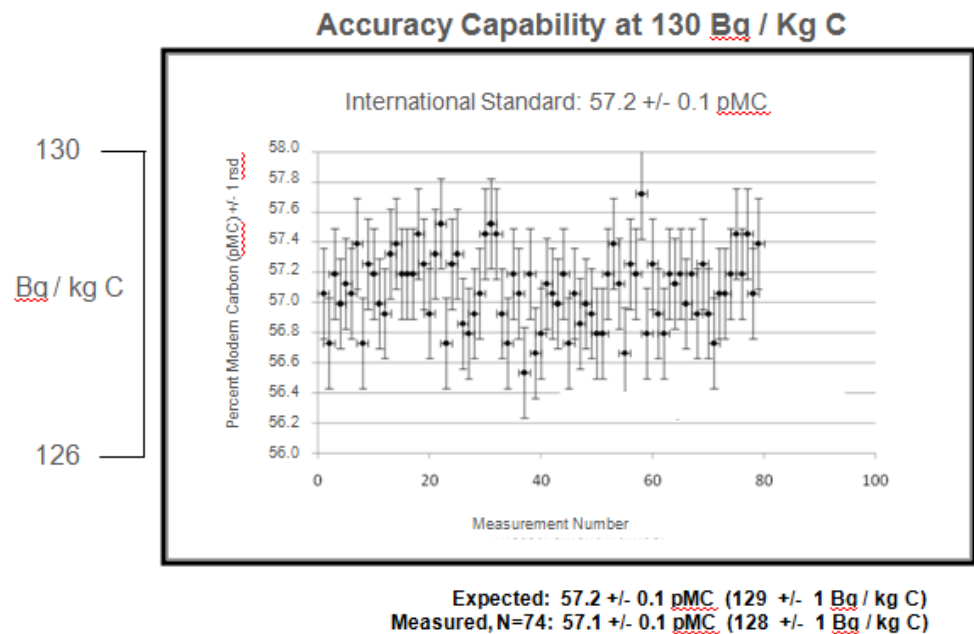


Figure 3-2  
Example of AMS Accuracy Capability (Provided by Beta Analytic, Inc.)







## Section 4: Radiocarbon in the Environment and Environmental Models

### 4.1 Historical and Anthropogenic Contributions to Radiocarbon

It is well documented that atmospheric production rates of carbon-14 ( $^{14}\text{C}$ ) vary with variations in cosmic flux influenced by solar winds and the earth's geomagnetic field. Long term variations are detectable in tree rings and ancient corals. These ancient records along with reservoir modeling suggest a global natural  $^{14}\text{C}$  inventory of about  $5 \times 10^4$  kilograms. (Key et.al., 2001) A recent production estimate for  $^{14}\text{C}$  from the reaction ( $^{14}\text{N}$  (n, p)  $^{14}\text{C}$ ) from natural sources is  $1.54 \text{ PBq/yr}^8$  (Argonne National Laboratory, 2005). Another earlier estimation of the production rate can be found in The Radioactivity of the Atmosphere and Hydrosphere (Suess, 1958) where the production was estimated at  $2.5 \text{ atoms/sec} \cdot \text{cm}^2$  multiplied by the earth's surface area of  $5.18 \text{ cm}^2$ , this production rate results in a flux of  $42,000 \text{ Ci/year}$  (or  $1.55 \text{ PBq/year}$ ).

Anthropogenic influences on atmospheric  $^{14}\text{CO}_2$  are well documented. Fossil  $\text{CO}_2$  influx with the onset of the industrial revolution is measurable after about AD 1880. By 1950, tree ring data suggests fossil fuel  $\text{CO}_2$  diluted the air  $^{14}\text{CO}_2$  signature<sup>9</sup> by approximately 3 percent modern carbon relative to natural production.  $^{14}\text{CO}_2$  influx from thermo-nuclear weapons testing is measurable after about AD 1959. Measurement of annual growth medium (plant and animal) indicates “bomb” carbon from weapons testing almost doubled the air  $^{14}\text{CO}_2$  signature by AD 1963.

Many data sets have been constructed based on bomb  $^{14}\text{C}$  data derived from atmospheric samples, tree rings, and organic materials; see Hua and Barbetti 2004 for a listing of studies (Hua & Barbetti, 2004; Wang et al., 2012). It has been estimated that  $2.13 \times 10^{17} \text{ Bq}$  (or  $5.76 \times 10^6 \text{ Curies}$ ) of  $^{14}\text{C}$  were released into the atmosphere by nuclear weapons testing (United Nations Scientific Committee on the Effects of Atomic Radiation, 2000). A more recent estimate suggests a higher value of  $3.55 \times 10^{17} \text{ Bq}$  (or  $9.59 \times 10^6 \text{ Curies}$ ) (Argonne National

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<sup>8</sup> Note that the SI unit prefix P (or peta) indicates a multiplier of  $1 \times 10^{15}$  and not to be confused with p (or pico) indicating a multiplier of  $1 \times 10^{-12}$ .

<sup>9</sup> The term signature refers to the values of the of the Fraction of Modern Carbon, Percent Modern Carbon, or Delta C-14 (see Equation 3-2, Equation 3-3, and Section 3.2).

Laboratory, 2005). 1963 marked the signing of the Partial Test Ban Treaty (PTBT) between the United States, United Kingdom and the Soviet Union wherein it was agreed nuclear weapons testing would no longer be performed above ground surface. Since that time, the atmospheric  $^{14}\text{CO}_2$  signature has declined to a present day level of approximately eight percent higher than pre-bomb era as illustrated in Figure 4-2.

The following discussion addresses the degree of atmospheric mixing over a several year period associated with atmospheric nuclear weapons testing. The  $^{14}\text{C}$  associated with weapons testing distributed itself un-evenly around the troposphere according to meteorological patterns and the fact that most atmospheric testing was conducted in the northern hemisphere. Studies were conducted within several zones within the world's troposphere as displayed in Figure 4-1, Zones For Monitoring  $^{14}\text{C}$  During and Following Weapons Testing. As expected the initial indications for the northern hemisphere, zones 1 and 2, nearly doubled in the percent modern carbon as observed in Figure 4-2. Within a few years of the cessation of testing,  $^{14}\text{C}$  declined and the distribution of  $^{14}\text{C}$  equilibrated over the zones as the excess  $^{14}\text{C}$  was evenly distributed throughout the troposphere and continually sequestered into oceans and biological systems (Hua and Barbetti, 2004).

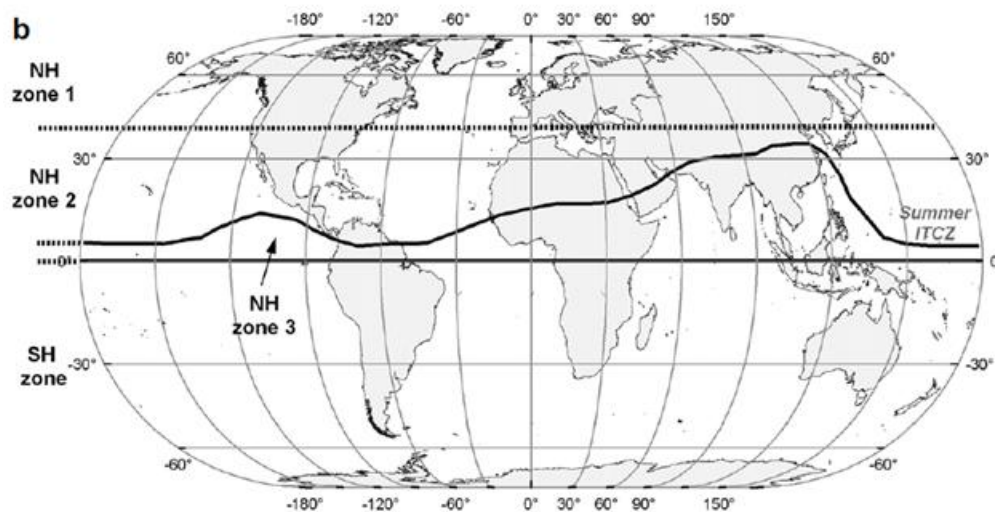


Figure 4-1  
Zones for Monitoring  $^{14}\text{C}$  during and Following Weapons Testing (Hua and Barbetti, 2004 and Hua et al., 2013; Creative Commons Attribution 3.0 License, <http://creativecommons.org/licenses/by/3.0/>)

Localized air  $^{14}\text{CO}_2$  signature is common, depending upon proximity to fossil fuel sources, nuclear power plants, and geothermal hot-spots. This effect is termed the “reservoir effect” and will be represented in localized plants and subsequent food chain members. Regional variations are also noted in the literature. In the United States, regional “zones” of difference are determined by comparing measured carbon-14 values in corn leaves to clear air measured in Pt. Barrow, Alaska [The unit of measure is  $\Delta^{14}\text{C}$  in per mil (o/oo), 10 per mil = 1

pMC, see Section 3 for a discussion of this unit] (Pataki et.al., 2010). As an example, the value of -22.3 per mil value noted in Southern California indicates air  $^{14}\text{CO}_2$  signature in Southern California is depleted in 2.2 pMC lower than air in Alaska. As such, if ambient is universally taken as 105 pMC, then this data suggests ambient for S. California is closer to 103.8 pMC.

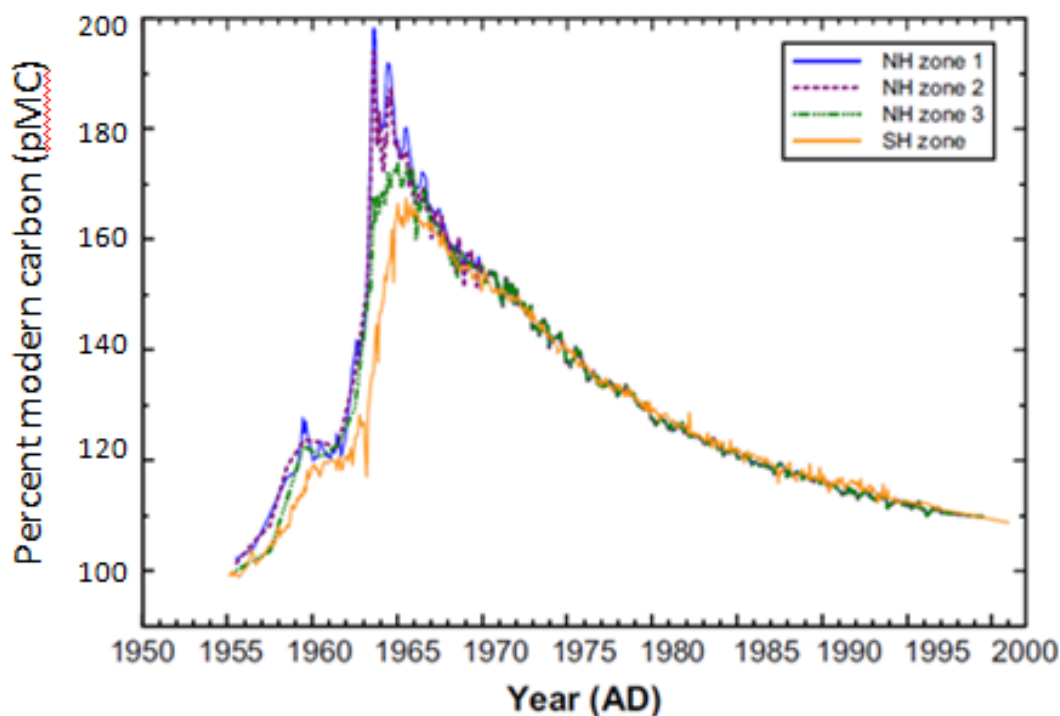


Figure 4-2

*Percent Modern Carbon in the Troposphere during and Following Weapons Testing (Hua and Barbetti, 2004; Creative Commons Attribution 3.0 License, <http://creativecommons.org/licenses/by/3.0/>)*

The “bomb curve” is a tool available to immediately assess the presence or absence of  $^{14}\text{CO}_2$  from anthropogenic influences including the effluents from nuclear generating stations. An ambient value (as of 2012) was approximately 105 pMC. Values greater than 105 pMC measured on plant samples which are harvested within areas affected by NPP  $^{14}\text{C}$  demonstrate: (1) uptake of NPP  $^{14}\text{C}$  and (2) the relative significance compared to levels within the atmosphere over the last 50 years.

Local variations in  $^{14}\text{CO}_2$  Signatures are well known to exist and have been mapped for certain geographic regions. For example, industrial areas of Southern California have been measured to be about 1 – 3 pMC depleted in  $^{14}\text{CO}_2$  relative to non-industrial areas such as northwest Washington State and central Minnesota (Pataki et.al, 2010). It is therefore necessary to factor out this variable by analyzing local control samples which are un-impacted by the facility under study should be measured in addition to the areas of potential impact (indicator

stations or samples). The difference between the two will best represent the impact from NPP  $^{14}\text{CO}_2$ .

### 4.1.1 Carbon-14 Inventories

Although carbon-14 ( $^{14}\text{C}$ ) is formed in the upper atmosphere it is distributed in the atmosphere, terrestrial, and aquatic compartments. A compilation is provided below in Table 4-1 of available data on quantities of carbon or carbon-14 present in the environment with the associated reference. The world's oceans contain the largest amount of carbon with 38,000 Pg<sup>10</sup>, followed by terrestrial with 2,200 Pg, and the atmosphere with 730 Pg (Matsumoto & Key, 2004). The oceans are the greatest repository of carbon and  $^{14}\text{C}$ . It is important to understand global  $^{14}\text{C}$  inventories because the total amount of carbon and  $^{14}\text{C}$  in the atmosphere impact how  $^{14}\text{C}$  will be incorporated into the human dose pathways. The upcoming portions of Section 4 address more detail regarding observations, trends, and models of these various environmental compartments.

*Table 4-1  
Carbon-14 and Carbon Inventories in Various Compartments/Media*

<b>Inventory Compartment</b>	<b>Quantity</b>	<b>Reference</b>
Atmospheric Inventory of $^{14}\text{C}$	13E+6 Ci (4.81E+17)	(Argonne National Laboratory, 2005)
Troposphere Concentration of $^{14}\text{C}$	3.4 pCi/kg air (0.126 Bq/kg air)	(Argonne National Laboratory, 2005)
Soil Concentration of $^{14}\text{C}$	0.2 pCi/g (7.4E-3 Bq/g)	(Argonne National Laboratory, 2005)
Total $^{14}\text{C}$ in Adult Human	~0.1 $\mu\text{Ci}$ (3.7E+3 Bq)	(Argonne National Laboratory, 2005)
Total Steady State Inventory of $^{14}\text{C}$	300 E+6 Ci (1.11E+19 Bq)	(Argonne National Laboratory, 2005)
World Ocean Inventory of Carbon	38,000 Petagram Carbon (Pg C)	(Matsumoto & Key, 2004)
Atmospheric Reservoir of Carbon	730 Pg C	(Matsumoto & Key, 2004)
Terrestrial Reservoir of Carbon	2200 Pg C	(Matsumoto & Key, 2004)

<sup>10</sup> Note that the SI unit prefix P (or peta) indicates a multiplier of  $1 \times 10^{15}$  and not to be confused with p (or pico) indicating a multiplier of  $1 \times 10^{-12}$ .

## **4.2 Current Models for Radionuclide Transport and Distribution in the Environment**

The ever-increasing use of nuclear materials has resulted in the development of numerous mathematical models to describe the transport of radionuclides in the environment. Radionuclides can enter the environment from a wide variety of sources: from surface and groundwater to soils, to dry deposition from air-borne releases, to foliar deposition on leaves from rainwater (Thiessen et.al., 1999).

It is relevant to mention that all models describing  $^{14}\text{C}$  transfer in the environment are based on the principle of isotope equilibrium, i.e., the specific activity of carbon (Bq of  $^{14}\text{C}$  per kg C) in plants is identical to the atmospheric specific activity for all trophic levels (Roussel-Debet et al., 2006). The specific activity of atmospheric carbon can then be calculated using the specific activity in plants that are in equilibrium with the surrounding medium, most notably by using tree rings (Roussel-Debet et al., 2006; Wang et al., 2012; Dias et al., 2008). Changes in the specific activity value have been observed over time- first an increase in the late 1950's to early 60's from weapons testing, followed by a decrease after the 1960's due to the exchange of atmospheric  $^{14}\text{CO}_2$  with oceanic carbonates and the continuous input of  $^{14}\text{C}$  depleted carbon from fossil fuels, as described Section 4.1 above.

### **4.2.1 Specific Activity and Compartment Models, Including Assumptions**

The specific activity of a radionuclide is defined as the activity per unit mass of radionuclide to its corresponding stable element. It has been noted since the early-1950s that the specific activity of carbon-14 in living organisms is in equilibrium with atmospheric levels. There is a deviation from this state due to dilution by stable  $\text{CO}_2$ , primarily from the burning of fossil fuels, in what is known as the Suess effect (Arnold & Anderson, 1957). The Suess effect is discussed in detail in Section 4.3.1. It represents a significant change in dose received from carbon-14.

#### **4.2.1.1 Two Approaches for Determining Model Parameters**

There are two approaches for determining carbon-14 model parameters. The first approach is based on element partitioning and accumulation concepts, which are qualitatively based on radionuclide transfer factors that describe transport between various environmental compartments (IAEA, 2010; IAEA, 2003). This approach is appropriate for most radionuclides as it accounts for factors such as bioaccumulation and transfer between compartments that exhibit time dependence.

The second approach is the specific activity method. Under equilibrium conditions (such as chronic release scenarios), the specific activity method provides an alternative to the multi-compartment model that requires the use of transport factors, and can be applied to long-lived isotopes of biologically regulated, essential elements that are highly mobile in the environment. Specific

activity models are commonly used to describe the dynamics of hydrogen ( $^3\text{H}$ ), carbon ( $^{14}\text{C}$ ), and chlorine ( $^{36}\text{Cl}$ ). The specific activity model assumes that the radioisotope of interest mixes physically and chemically with its corresponding stable element within each environmental compartment, thus resulting in a constant specific activity level. An organism that draws from this environmental compartment will attain the same specific activity as the source compartment.

Specific activity models are frequently used by regulators because they result in conservative estimates as complete equilibrium is unlikely to be obtained in real environmental compartments (Galeriu et al., 2007; Wirth, 1982). For the United States Nuclear Regulatory Commission (US NRC), this means that the concentration of carbon-14 in vegetation is calculated by assuming that its ratio to total carbon in the atmosphere around the vegetation is the same, and subsequently animal concentrations are calculated using concentration factors and animal intake rates (Wirth, 1982; US NRC, 1977). Then the dose rate to man from ingestion of contaminated foodstuffs is proportional to his intake rate of the foodstuffs and the dose commitment factor (DCF) for carbon-14.

#### 4.2.1.2 Validity of Specific Activity Models for Carbon-14

The majority of carbon-14 released to atmosphere by nuclear power plants is emitted as  $^{14}\text{CO}_2$  (Wirth, 1982; NCRP, 1985). The assumption of full specific activity equilibrium throughout the terrestrial environment is completely satisfactory for atmospheric releases of carbon-14 if it is emitted as  $^{14}\text{CO}_2$  (IAEA, 2010). For the specific activity model to be valid, the relation between  $^{14}\text{C}$  and  $^{12}\text{C}$  must be constant from the atmosphere throughout the entire food chain. A shift in this relation can only occur via isotopic effects (Wirth, 1982). Isotopic exchange with uncontaminated pools of the stable element can result in the progressive dilution of the isotope as a function of distance from the source compartment. The results of the study by Wirth in 1982 show that the specific activity of carbon-14 in man follows that in air, and thus the specific activity model is suitable for dose calculations for  $^{14}\text{C}$  (Wirth, 1982). The radiation exposure will be overestimated by assuming that all  $^{14}\text{C}$  is released as  $^{14}\text{CO}_2$ .

Killough & Rohwer (1978) performed an analysis of the dosimetry of  $^{14}\text{C}$  released to the atmosphere from nuclear power plants as  $^{14}\text{CO}_2$  (Killough & Rohwer, 1978). Their results suggested that dose rate factors based on the specific activity model were appropriate in so far as exposures were dominated by ingestion, a reasonable assumption according to Galeriu et al, Key, and Takahashi (Galeriu et al., 2007; Key, n.d.; Takahashi et al., 2011). Killough & Rohwer did note, however, that because of the photosynthetic link in the food chain to man, there was need for adjustments to meteorological frequencies in the atmospheric dispersion model, and they suggested a renormalization such that the specific activity of dietary carbon would reflect average ground level air concentrations ( $\chi$ ) for daylight growing season hours (Killough & Rohwer, 1978). For additional discussion refer to Section 6.5.

#### 4.2.1.3 The Widespread Usage of Specific Activity Models

The constancy of carbon-14 specific activity in living organisms and in atmosphere has resulted in the widespread use of specific activity assumptions in models for dose calculations. The IAEA utilizes a specific activity model for carbon-14, consistent with literature (Sheppard et al., 2006; Wirth, 1982; Sheppard et al., 2006; Keum et al., 2008; Avila & Prohl, 2008; Xu et al., 2011; Galeriu et al., 2007; Killough & Rohwer, 1978; IAEA, 2010).

The IAEA uses a simple equation to calculate the carbon-14 content in fresh weight plants due to releases of carbon-14 to the atmosphere:

$$C_{p_{fw}} = \frac{C_{air} * S_p}{S_{air}} \quad \text{Eq. 4-1}$$

Where:

$C_{p_{fw}}$  is carbon-14 concentration in the fresh weight plant (Bequerel per kilogram fresh weight, Bq kg<sup>-1</sup>)

$S_p$  is the concentration of stable carbon in the plant (gram carbon per kilogram fresh weight, gC kg<sup>-1</sup>)

$C_{air}$  is the concentration of carbon-14 in air (Bequerel per cubic meter, Bq m<sup>-3</sup>, known via measurements or models)

$S_{air}$  is the stable carbon concentration in air (gram carbon per cubic meter of air, gC m<sup>-3</sup>), taken to be 0.20 g m<sup>-3</sup> (IAEA 2010).

Table 4-2 below, reproduced from IAEA Technical Report Series No. 472 shows measured values of the carbon content for various plant categories.

Table 4-2

*Concentration of Stable Carbon in Terrestrial Plants, Table 66 in IAEA Handbook 472 (IAEA, 2010)*

<b>Stable Carbon Content in Terrestrial Plants (gC kg<sup>-1</sup> fresh weight)</b>		
<b>Plant Category</b>	<b>Average</b>	<b>Maximum</b>
<b>Leafy and non-leafy vegetables</b>	<b>3.0 × 10<sup>1</sup></b>	<b>6.5 × 10<sup>1</sup></b>
<b>Leguminous vegetables</b>		
Seed	4.1 × 10 <sup>2</sup>	4.7 × 10 <sup>2</sup>
Vegetative mass	5.9 × 10 <sup>1</sup>	1.1 × 10 <sup>2</sup>
<b>Root crops</b>	4.6 × 10 <sup>1</sup>	9.5 × 10 <sup>1</sup>
<b>Tubers<sup>11</sup></b>	10.3 × 10 <sup>1</sup>	1.3 × 10 <sup>1</sup>
<b>Fruit</b>	6.2 × 10 <sup>1</sup>	1.0 × 10 <sup>2</sup>
<b>Grass, fodder, pasture</b>	1.0 × 10 <sup>2</sup>	1.6 × 10 <sup>2</sup>
<b>Cereals (including rice)</b>	3.9 × 10 <sup>2</sup>	4.3 × 10 <sup>2</sup>
<b>Maize</b>		
Sweet corn	1.2 × 10 <sup>2</sup>	1.2 × 10 <sup>2</sup>
Feed corn	3.8 × 10 <sup>2</sup>	–
<b>Silage</b>	1.3 × 10 <sup>2</sup>	1.8 × 10 <sup>2</sup>

The equation for carbon-14 concentration in animal products is similar to that for plants:

$$C_{afw} = \frac{f_a * C_{pfw} * S_a}{S_p} \quad \text{Eq. 4-2}$$

<sup>11</sup> The value provided in Table 66 of the IAEA Handbook for tubers is 10.3 × 10<sup>2</sup> g of C per kg fresh weight which is not possible; this is an error in the decimal placement or exponent. The correct value is 1.03 × 10<sup>2</sup> g of C per kg fresh weight.



Where:

$C_{afw}$  is the carbon-14 concentration in animal products (Bq kg<sup>-1</sup> fresh weight)

$f_a$  is the fraction of animal feed that is contaminated

$S_a$  is the concentration of stable carbon in the animal product (gC kg<sup>-1</sup> fresh weight) (IAEA 2010).

Note that the  $f_a$  value allows for animals to be fed from remote sources that are not impacted by nuclear power plant effluents, and the value used should be set based on consideration of local farming practices. If no site-specific values are available,  $f_a$  should be conservatively set at unity. Table 4-3 below shows the stable carbon content of various animal products.

Table 4-3

Concentration of Stable Carbon in Terrestrial Animal Products, Table 67 in IAEA Handbook 472 (IAEA, 2010)

Stable Carbon Content in Terrestrial Animal Products (gC kg <sup>-1</sup> fresh weight)		
Animal Product	Average	Maximum
<b>Milk</b>		
Cow	$6.5 \times 10^1$	$6.9 \times 10^1$
Sheep	$1.1 \times 10^2$	–
Goat	$7.1 \times 10^1$	–
<b>Meat</b>		
Beef	$2.0 \times 10^2$	$2.9 \times 10^2$
Veal	$1.6 \times 10^2$	$1.9 \times 10^2$
Mutton	$2.9 \times 10^2$	–
Lamb	$2.8 \times 10^2$	$3.2 \times 10^2$
Goat	$1.7 \times 10^2$	–
Pork	$3.0 \times 10^2$	$5.5 \times 10^2$
Hen	$2.4 \times 10^2$	–
Broiler	$1.5 \times 10^2$	$2.0 \times 10^2$
<b>Egg</b>	$1.6 \times 10^2$	$1.6 \times 10^2$

The US NRC also uses the specific activity model in its guidance for calculating carbon-14 concentrations in vegetation. Specific guidance is provided for calculation of carbon-14 in vegetation in Regulatory Guide 1.109, “Calculation of Annual Doses to Man from Routine Releases of Reactor Effluents for the Purpose of Evaluating Compliance with 10 CFR Part 50, Appendix I” (USNRC 1977b). Vegetation, in this context includes, forage, produce, and leafy vegetation.

$$C_{14}^V(r, \theta) = 3.17 \times 10^7 p Q_{14} \left[ \frac{x}{Q} \right] (r, \theta) 0.11/0.16 \quad \text{Eq. 4-3}$$

(note this is equation C-8 Regulatory Guide 1.109 (USNRC, 1997b))

Or

$$C_{14}^V(r, \theta) = 2.2 \times 10^7 p Q_{14} \left[ \frac{x}{Q} \right] (r, \theta) \quad \text{Eq. 4-4}$$

Where:

$C_{14}^V(r, \theta)$  is the concentration of carbon-14 in vegetation grown at location  $(r, \theta)$  in pCi/kg

$\left[ \frac{x}{Q} \right] (r, \theta)$  is the annual average atmospheric dispersion factor in the sector at angle  $\theta$  at distance  $r$  from the release point in  $\text{sec/m}^3$

$Q_{14}$  is the annual release rate of carbon-14 in Ci/yr, Note that “C-14 is assumed to be released in oxide form (CO or CO<sub>2</sub>)” for this equation in Regulatory Guide 1.109) (USNRC, 1977b)

$p$  is the fractional equilibrium ratio, dimensionless, “Also in the case of intermittent releases such as from gaseous waste decay tanks the parameter  $p$  is employed to account for the fractional equilibrium ratio.  $p$  is defined as the ratio of the total annual release time (for C-14 atmospheric releases) to the total annual time during which photosynthesis occurs (taken to be 4400 hours). Under this condition the value of  $p$  should never exceed unity. For continuous C-14 releases,  $p$  is taken to be unity.” (USNRC, 1977b)

0.11 is the total plant mass that is natural carbon, dimensionless

0.16 is equal to the concentration of natural carbon in the atmosphere ( $\text{gC/m}^3$ ), and

$3.17 \times 10^7$  is equal to  $(1.0 \times 10^{12} \text{ pCi/Ci})(1.0 \times 10^3 \text{ g/kg})/(3.15 \times 10^7 \text{ sec/yr})$

At this point, it is interesting to note the difference in the concentration of natural carbon in the atmosphere used by IAEA and the US NRC. IAEA uses a value of  $0.20 \text{ g m}^{-3}$  while the US NRC uses  $0.16 \text{ g m}^{-3}$  in RG 1.109. See Section 4.3.1 discussion of the Seuss effect for more details on this parameter. Furthermore, the impact of this difference is discussed in Section 6.

Specific activity models have been compared to more complex, dynamic models, and have produced consistent results and/or otherwise shown convergence with the specific activity models (Keum et al., 2008; Takahashi et al., 2011). Some models are based on specific activity considerations, but include a time-dependence in order to properly account for seasonal variations (Sheppard et al., 2006; Sheppard et al., 2006). Overall, specific activity models that incorporate a time-dependence factor are appropriate for regulatory uses, as they estimate doses in a realistic, yet conservative, manner.

#### **4.2.2 Transfer Mechanisms for Carbon in Plants**

There are three transfer mechanisms for carbon in plants: photosynthesis, respiration, and translocation. Photosynthesis represents the pathway by which radiolabeled  $\text{CO}_2$  is taken up by the plant. In photorespiration, plants release a small portion of the  $\text{CO}_2$  that was not utilized in photosynthetic processes. While respiration plays an important role in the dynamics of carbon-14 modeling, it likely represents only a small sink in the total amount of  $\text{CO}_2$  removed from the air by the plant. As there are no models that are able to accurately quantify  $^{14}\text{CO}_2$  removal via respiration, it is neglected in this analysis. Carbon can also be trans-located from plant roots to the edible portions of the plant. Translocation is primarily of concern in waste repository modeling, where highly contaminated soils are possible. For regular releases from nuclear power plants, translocation can be considered negligible (Amiro et al., 1991).

Atmospheric  $^{14}\text{CO}_2$  is taken into the plant and transformed into organic matter through photosynthesis. This organic matter accumulates in each portion of the plant at each growing stage, and some of the  $^{14}\text{CO}_2$  is lost back to the atmosphere via respiration, resulting in the cyclical nature of carbon. The primary pathway for the incorporation of  $^{14}\text{CO}_2$  into plants is via the leaf by photosynthesis of  $\text{CO}_2$  including  $^{14}\text{CO}_2$  (Roussel-Debet et al., 2006; Takahashi et al., 2011; Amiro et al., 1991).

Plants that grow in soils containing  $^{14}\text{C}$  can accumulate  $^{14}\text{C}$  from direct uptake through the roots and from indirect uptake from the atmosphere (Amiro et al., 1991). There are several factors that influence the atmospheric contribution to the plant from the soil: volatilization rate from the soil; source surface area; and meteorological dispersal conditions. It is relevant to note that as only 1-2% of plant carbon is absorbed through the roots, this pathway is essentially negligible. (Amiro et al., 1991; Sheppard et. al., 1991).

Figure 4-3, Conceptual Illustration of a Compartment Model for carbon-14 Atmospheric Releases, is an illustration of a compartment model for atmospheric releases of  $^{14}\text{C}$  as  $^{14}\text{CO}_2$  from nuclear facilities, derived from Takahashi et. al. (Takahashi et. al., 2011).

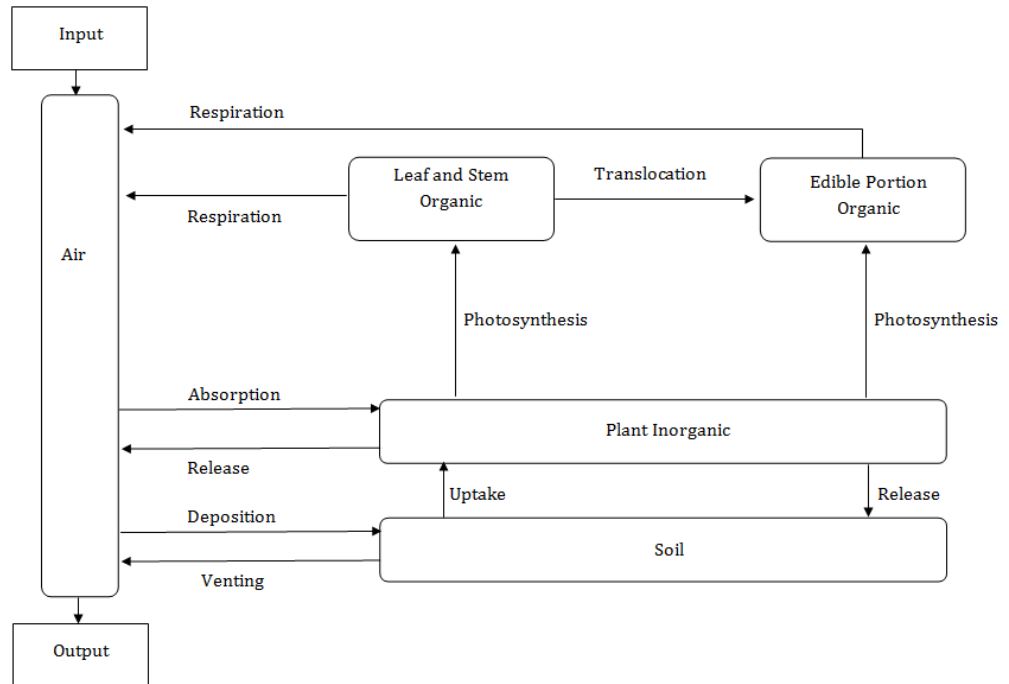


Figure 4-3  
Conceptual Illustration of a Compartment Model for Carbon-14 Atmospheric Releases (Derived from Takahashi et. al., 2011)

#### 4.2.2.1 Photorespiration

Fisher studied the ecosystem respiration as a function of precipitation and heat stress in Spatiotemporal Variations in Growing Season Exchanges of  $\text{CO}_2$ ,  $\text{H}_2\text{O}$ , and Sensible Heat in Agricultural Fields of the Southern Great Plains (Fisher et al., 2007). During dry conditions, plant stomas<sup>12</sup> close to conserve moisture, ceasing  $\text{CO}_2$  intake. Initial carbon fixation occurs in the chloroplast. When  $\text{CO}_2$  saturation occurs, typically at midday, the plant ceases to assimilate  $\text{CO}_2$  and may result in some  $\text{CO}_2$  being released via photorespiration. It is possible that photorespiration plays an important role in the dynamics of  $^{14}\text{C}$  cycling. However there are currently no models that can accurately quantify the contribution from respiration (Key, no date).

#### 4.2.3 The Ocean: An Exchange Reservoir for Carbon

It is well known that the world's oceans serve as the exchange reservoir in the carbon cycle. Figure 4-4 provides a schematic of the model of carbon in the ocean. Dissolved  $\text{CO}_2$  is found in three main forms:  $\text{CO}_2$ ,  $\text{CO}_3^{2-}$ , and  $\text{HCO}_3^-$ , the sum of which is termed dissolved inorganic carbon, or DIC. DIC is transported within the ocean by both physiological and biological processes. Gross primary production, or GPP, is the total amount of organic carbon formed

<sup>12</sup> **Stoma** is a pore, found in the epidermis of leaves, stems and other organs that is used to control gas exchange.

by photosynthesis; net primary production is the organic carbon that remains after respiration by photosynthetic organisms. Dissolved organic carbon (DOC) and particulate organic matter (POC) are transpired and respired by non-photosynthetic organisms, and this is ultimately upwelled and returned to atmosphere, with only a tiny fraction getting buried in deep-sea sediments as  $\text{CaCO}_3$ . The fluxes (values provided in PgC/yr) shown in the schematic in Figure 4-4 are for the short term burial of both organic carbon and  $\text{CaCO}_3$  in coastal sediments (Prentice et al., 2001). The dashed lines represent fluxes of carbon as  $\text{CaCO}_3$ , the solid lines represent organic carbon.

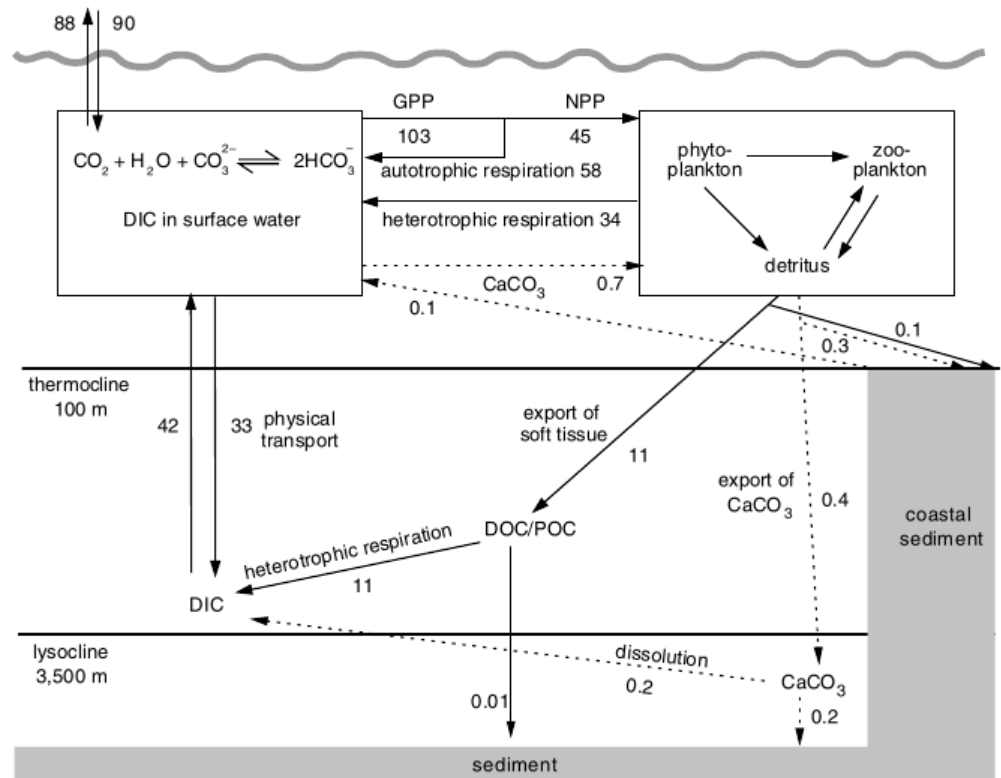


Figure 4-4  
Carbon Cycling in the Ocean (Prentice et.al., 2001; Page 188, Figure 3-1c)

Several papers have been published (Kumblad et al., 2003; Sheppard et al., 2006) that seek to create ecosystem level models for the transport of  $^{14}\text{C}$  in the ocean, however these model are relatively site specific. In general, for surface waters and scenarios where there is fast water exchange (e.g., in a bay),  $^{14}\text{C}$  is quickly diluted with only small levels assimilating into the food chain. For deep ocean models, levels of  $^{14}\text{C}$  available for uptake deviate significantly from each other on both a spatial scale and a temporal scale, depending on which ventilation model is used (Matsumoto & Key, 2004). There is no single model that can adequately describe the movement of  $^{14}\text{C}$  in the ocean environment.

### 4.3 Summary and Discussion of Atmospheric Carbon-14 and Trends

The abundance of atmospheric CO<sub>2</sub> is ultimately controlled by exchange with the organic and inorganic carbon reservoirs, namely Earth's oceans. Decreasing levels of <sup>14</sup>CO<sub>2</sub> up to the 1990's were governed by the equilibration of atmospheric bomb <sup>14</sup>C perturbation with oceans and the terrestrial biosphere. The largest perturbations today are the emissions of fossil fuel CO<sub>2</sub> that are depleted of <sup>14</sup>C. Fossil fuel use dilutes global atmospheric <sup>14</sup>CO<sub>2</sub> by 12-14% per year, and this loss is only partially compensated by <sup>14</sup>CO<sub>2</sub> releases from the biosphere, industrial emissions, and natural production in the upper atmosphere (Levin et al., 2010).

#### 4.3.1 Suess Effect, Trends in CO<sub>2</sub> Releases

The dilution of <sup>14</sup>CO<sub>2</sub> with plain CO<sub>2</sub> was first noted by Hans Suess in 1955 (Suess, 1955). At the time the cause of this dilution was unclear, and several theories were hypothesized. It is now well known and accepted that the combustion of fossil fuels continually reduces the level of radio-labeled carbon dioxide in the atmosphere (Arnold & Anderson, 1957; NCRP, 1985; Craig, 1957; Hua, 2009; Yim & Caron, 2006; Prentice et al., 2001; Collins & Bell, 2001). Urban areas can have elevated CO<sub>2</sub> levels from human emissions as well, reducing the long-term impact of carbon-14 on those populations (Key, n.d.).

Much effort has gone into quantifying the decrease in the atmospheric <sup>14</sup>C as a result of the dilution, primarily because of the impact on radiocarbon dating. Regulatory guide 1.109 (1977) recommends a value of 0.16 gC m<sup>-3</sup> (based on an assessment of CO<sub>2</sub> concentration of 326 ppm CO<sub>2</sub> in 1977<sup>13</sup>) for the stable carbon content of air, while the IAEA (2010) has adjusted this value to 0.20 gC m<sup>-3</sup> (based on a current as of 2010 global inventory of 383 ppm CO<sub>2</sub>) (USNRC, 1977; IAEA, 2010). The value used by the IAEA is already somewhat out of date as the current level of CO<sub>2</sub> is 395 ppm as of January 2013, showing an increase of 3 ppm over the last year alone (Dlugokencky & Tans, 2013).

As reliance on fossil fuels continues to increase, so does the level of stable CO<sub>2</sub> in the atmosphere. Figure 4-5 shows recent monthly mean carbon dioxide concentrations globally averaged over marine surface sites (Dlugokencky & Tans, 2013). The red line represents monthly mean values centered on the middle of each month; the black line represents the same, but corrected for average seasonal cycle. The correction is determined as a moving average of seven adjacent seasonal cycles centered on the month to be corrected, except for the first and last three and one-half years of the record, where the seasonal cycle has been averaged over the first and last seven years, respectively (Dlugokencky & Tans, 2013). The Global Monitoring Division of the National Oceanic and Atmospheric Administration's Earth System Research Laboratory (NOAA/ESRL) has measured carbon dioxide and other greenhouse gases

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<sup>13</sup> Note: This is the date of publication of the U.S. Nuclear Regulatory Commission Regulatory Guide 1.109 (U.S. Nuclear Regulatory Commission, 1977b).

for several decades at a globally distributed network of air sampling sites (Conway et al., 1994).

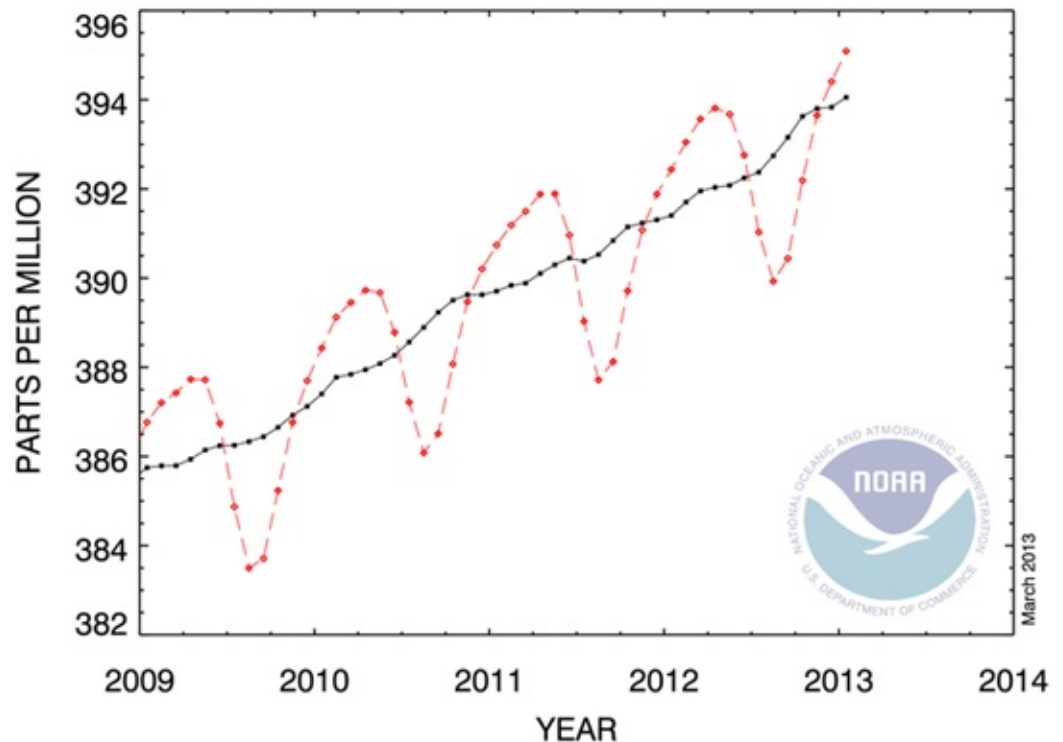


Figure 4-5

Recent Global Monthly Mean  $\text{CO}_2$  from <http://www.esrl.noaa.gov/gmd/ccgg/trends/global.html> (Dlugokencky & Tans, 2013)

The Mauna Loa Observatory in Hawaii has the longest continuous record of direct atmospheric  $\text{CO}_2$  measurements, with data dating back to 1958 (Tans & Keeling, 2013). Figure 4-6 shows all atmospheric  $\text{CO}_2$  concentrations recorded at the Mauna Loa Observatory. The red curve is the direct carbon dioxide data measured as the mole fraction in dry air; the black curve represents the seasonally corrected data (correction described above). Note that the observations shown are from both the Scripps Institute of Oceanography (starting March 1958), and have been independently measured and verified by the National Oceanic and Atmospheric Administration starting in May of 1974 (Keeling et al., 1976; Thoning et al., 1989).

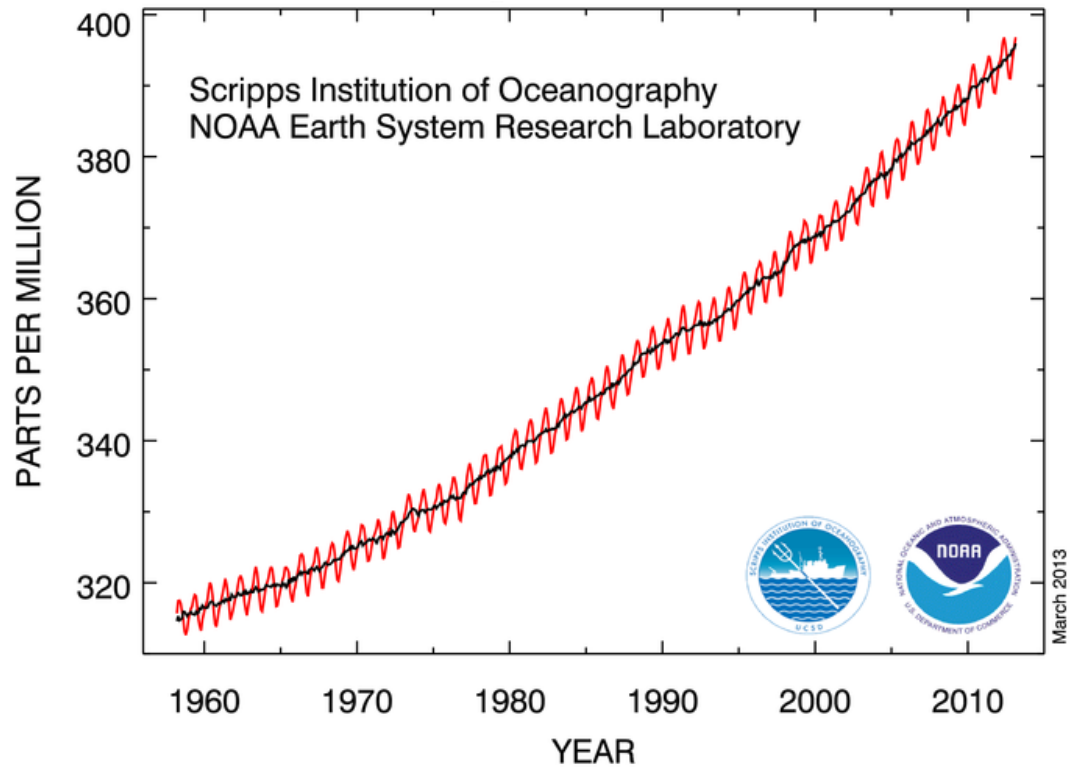


Figure 4-6  
 Atmospheric CO<sub>2</sub> Recorded at Mauna Loa Observatory from  
<http://www.esrl.noaa.gov/gmd/ccgg/trends/mlo.html> (Tans & Keeling, 2013)

Figure 4-7 shows the annual average carbon dioxide growth rate for the Mauna Loa Observatory. The black horizontal lines are decadal averages of the growth rate (Tans & Keeling, 2013). The annual mean growth rate of carbon dioxide is equivalent to the difference in concentration between the end of December and the start of January of the same year. If taken as a global average, it represents the sum of all carbon dioxide added to and removed from both human activities and natural processes. The estimated uncertainty in the Mauna Loa annual mean growth rate is 0.11 ppm yr<sup>-1</sup>, based on the standard deviation of the differences in the monthly mean values measured independently by the Scripps Institute of Oceanography and NOAA/ESRL (Tans & Keeling, 2013). While the global average growth rate and the growth rate seen at Mauna Loa are not the same, they are similar- one standard deviation of the annual differences at Mauna Loa Observatory minus the global average is 0.26 ppm yr<sup>-1</sup> (Tans & Keeling, 2013).



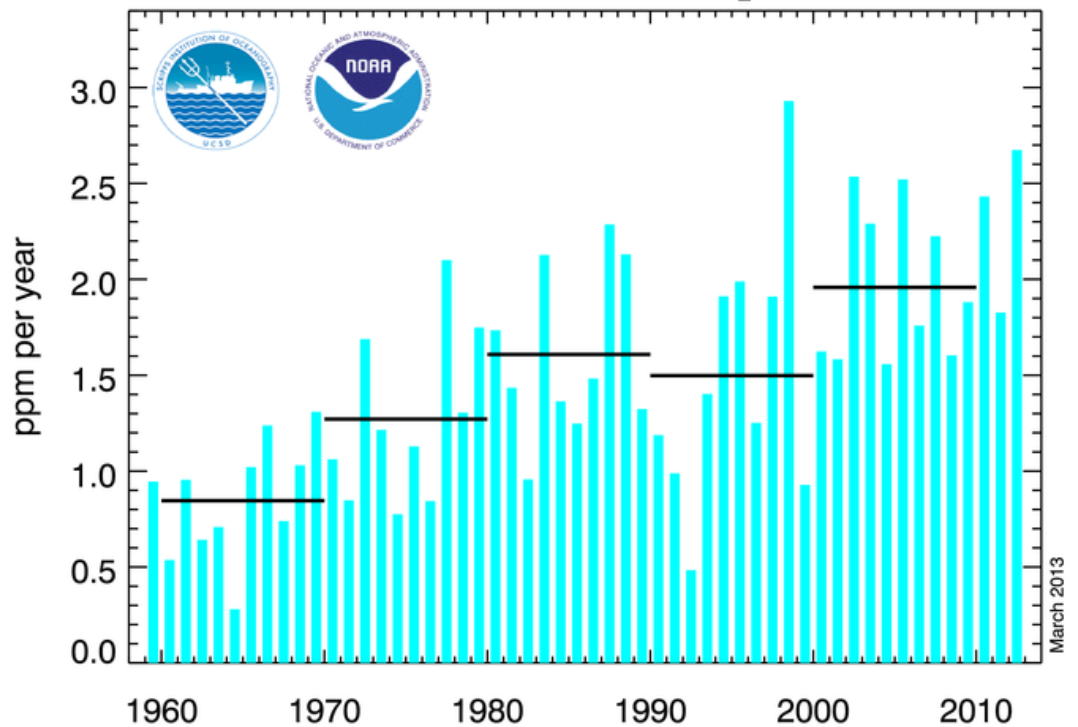


Figure 4-7

Annual Mean Growth Rate of CO<sub>2</sub> at Mauna Loa Observatory from  
<http://www.esrl.noaa.gov/gmd/ccgg/trends/mlo.html> (Tans & Keeling, 2013)

The increase in stable carbon dioxide levels is likely to continue as the demand for energy increases, and more fossil fuels are burned to meet that demand. For the purposes of this study, the implications of the increasing CO<sub>2</sub> levels are an increasing concentration of stable carbon; this ultimately results in a reduced dose impact from carbon-14.

#### 4.4 Transfer Coefficients

Transfer coefficients describe the uptake of radionuclides into a product (e.g. edible plants or animal milk) from a source. Transfer factors allow the activity concentrations in environmental media (e.g., soils, forage) to be directly related to the activity in plant and animal products (Thiessen et al., 1999). There are many types of transfer factors, relevant ones are defined here. The soil to plant transfer factor is the ratio of the dry weight concentration in the plant part of interest (e.g., edible portions) to the dry weight concentration in the specified soil layer (activity concentrations are calculated at standardized soil depths, 10 centimeters (cm) for grass, 20 cm for everything else). For animals, there are two types of transfer factors to consider: one is the forage to milk, the other is for forage to meat. The transfer of radionuclides from feed to milk, meat and other animal products is generally calculated using element-specific transfer factors, defined as the ratio of the activity in meat or milk to the daily activity intake under steady-state conditions (Thiessen et al., 1999). These transfer factors are then only valid under conditions of constant, long-term activity intake rates, an

assumption that is met for the regular releases of  $^{14}\text{C}$  from nuclear power plants. Table 4-4 below shows transfer factors for milk and meat products from various animals, as well as a transfer coefficient for eggs from various sources. Note that the IAEA does not recommend the use of transfer factors for carbon-14, rather they propose that the specific activity model (i.e. equilibrium concentrations in all compartments) is valid for all realistic cases (IAEA, 2010).

*Table 4-4*

*Comparison of Transfer Coefficients for  $^{14}\text{C}$  from various sources. CSA is Canadian Standards Association; GRG is the German Regulatory Authority. US NRC References: (Ng et al., 1968; Altman & Altman, 1968)*

<b>Product</b>	<b>RG 1.109 (US NRC, 1977)</b>	<b>CSA (taken from Galeriu et al., 2007)</b>	<b>GRG (taken from Galeriu et al., 2007)</b>	<b>Galeriu (Galeriu et al., 2007)</b>	<b>Limer et al. 2010 (Limer et al., 2010)</b>
<b>Milk (days per liter, d L<sup>-1</sup>)</b>					
Cow	$1.2 \times 10^2$	$1.5 \times 10^2$	$4.0 \times 10^2$	$1.1 \times 10^2$	$1 \times 10^2$
Sheep	–	–	–	$1.42 \times 10^1$	–
Goat	$1.0 \times 10^1$	–	–	$6.7 \times 10^2$	–
<b>Meat (days per kilogram, d kg<sup>-1</sup>)</b>					
Unspecified	$3.1 \times 10^2$	–	$2.0 \times 10^2$	–	–
Beef	–	$6.4 \times 10^2$	–	$4.6 \times 10^2$	$4 \times 10^2$
Veal	–	–	–	$8.5 \times 10^2$	–
Mutton	–	–	–	$3.96 \times 10^1$	–
Lamb	–	–	–	$4.19 \times 10^1$	–
Goat	–	–	–	$3.41 \times 10^1$	–
Pork	–	$1.8 \times 10^1$	–	$2.28 \times 10^1$	–
Hen	–	–	–	3.532	–
Poultry	–	4.2	–	–	–
Chicken	–	–	–	3.355	–
<b>Eggs (d kg<sup>-1</sup>)</b>	–	3.1	–	2.195	–

## 4.5 Models

The following presents carbon-14 models developed by European organizations to describe carbon-14 transport.

#### **4.5.1 TOCATTA and TOCATTA- $\chi$ Models**

The TOCATTA<sup>14</sup> models were developed by the French Institute for Radiological Protection and Nuclear Safety (IRSN) in France, under the collaborative network SYMBOISE. TOCATTA is a dynamic compartment model used to describe <sup>14</sup>C transfer in agricultural systems that are exposed to atmospheric carbon-14 releases from nuclear facilities under both normal and accident conditions (Le Dizès et al., 2012). This model has been verified against data taken over a two year period downwind of the La Hague nuclear fuel reprocessing plant. While TOCATTA roughly reproduces month-to-month variability, responses to accidental releases during the daytime are consistently under-estimated by as much as 33%, suggesting that the temporal resolution of the mode needs to be increased for it to be able to simulate intermittent daytime releases of <sup>14</sup>C. That said, the advantage of this model is its relative simplicity: the number of compartments and input parameters has been minimized where possible.

The TOCATTA model is based on a daily time-step, and is driven by atmospheric <sup>14</sup>CO<sub>2</sub> concentrations and meteorological data. Emissions as anything other than carbon dioxide are neglected, as they are not directly used in photosynthetic processes, and given its long half-life, radioactive decay is not considered.

The model contains five compartments: canopy atmosphere, soil water, soil air, soil organic matter, and plant dry material. Each compartment is described by specific transfer processes: volatilization, wet input to soil, total respiration, decomposition, and net primary production, respectively. See Le Dizès et al. 2012 for additional details (Le Dizès et al., 2012).

TOCATTA- $\chi$  is an update to the basic TOCATTA model, and it incorporates a pasture model for simulating grassland carbon and radiocarbon cycling called PaSiM. The TOCATTA- $\chi$  model has been benchmarked against observations of <sup>14</sup>C activity concentrations in the grassland around the La Hague reprocessing plant at two separate pastures, one intensively managed, with grass cut on a monthly basis and, the second, a poorly managed grass field. Results suggested that the impacts of land use (i.e. the impact of frequent cuts or grazing by cattle), may be stronger than the impact of meteorological conditions on the airborne radiocarbon uptake by grasses.

When atmospheric <sup>14</sup>C activity concentrations are directly derived from observational data, TOCATTA- $\chi$  performs well (Aulagnier et al., 2013). TOCATTA- $\chi$  performs well at reproducing general trends in grass carbon-14 activity concentrations, and is a useful tool for assessing doses induced by radioactive releases from nuclear facilities under both normal and accident operating conditions.

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<sup>14</sup> No available name for the model in literature.

#### **4.5.2 POM <sup>14</sup>C**

A process-oriented assessment model for uptake of carbon and doses from releases of carbon-14 to air was developed for use by the Swedish regulatory authority, and is called Process Oriented Model <sup>14</sup>C (POM<sup>14</sup>C). POM<sup>14</sup>C uses a model that describes the turnover of carbon in crops, and considers several possible exposure pathways, including: direct consumption of cereals, vegetables, and root vegetables grown near nuclear facilities, and milk and meat consumption from cows having eaten fodder or grazed on grasses grown around a nuclear power plant (Aquilonius & Hallberg, 2005).

POM<sup>14</sup>C calculations are performed in three distinct stages. First, the determination of carbon content of crop per unit area at harvest. This section of the model encompasses site-specific details of plant growth, including photosynthetic rate, growing season length, temperature, and global radiation. Next the derivation of the specific activity of carbon-14 in the foodstuff is calculated, assuming complete isotopic mixing (i.e., equilibrium) such that the ratio of carbon-14 concentration to stable carbon concentration in the foodstuff is the same as the ratio in air. To perform this calculation, the crop dependent yield and the carbon-14 activity in atmosphere are necessary. Finally, once the concentration of carbon-14 in the foodstuff is known, calculations to obtain dose to an individual member of the public is straightforward based on assumed exposure pathways (Xu et al., 2011). A diagram of the model is shown in Figure 4-8 below.

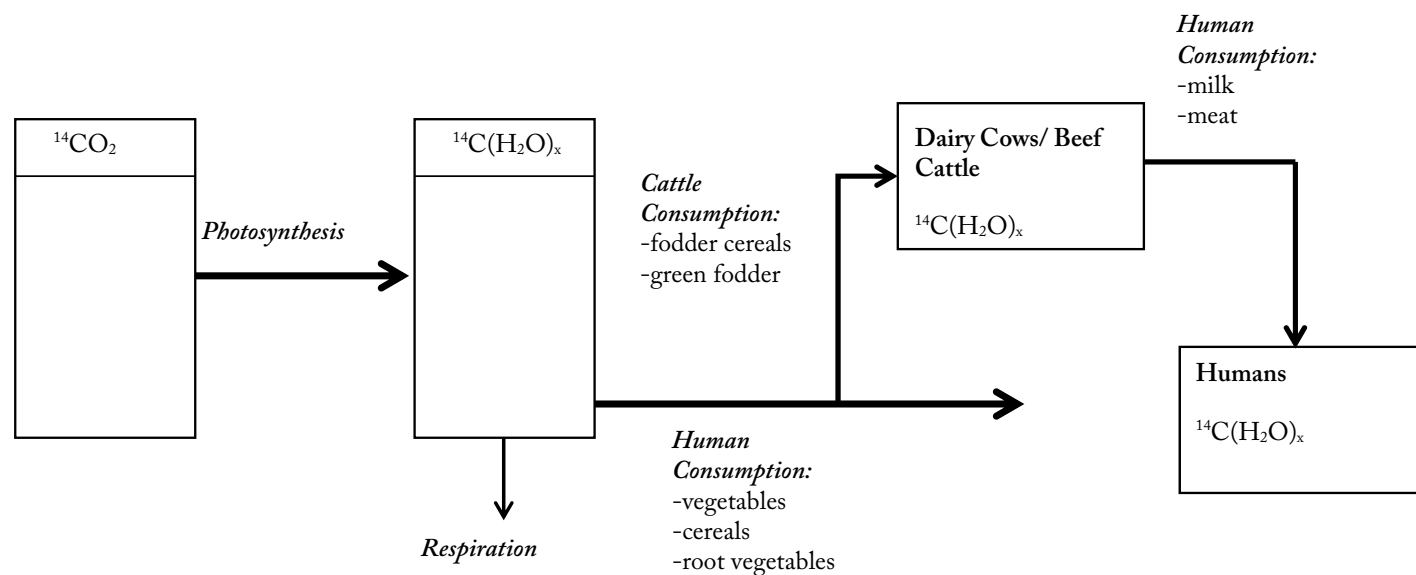


Figure 4-8  
Processes and Exposure Pathways in POM $^{14}\text{C}$  Model. (Adapted from Aquilonius and Hallberg, 2005.)

Some model assumptions include the following: all plants have an unlimited supply of CO<sub>2</sub> and water; carbon content at harvest is a function of several variables, values for which were taken to be seasonal averages; and the fractionation effect was disregarded, thus assuming that <sup>14</sup>C accumulates in vegetation at the same rate as stable carbon. More information and parameter values can be found in (Aquilonius & Hallberg, 2005; Xu et al., 2011).

#### **4.5.3 Model Summary**

The discussion of TOCATTA, TOCATTA- $\chi$ , and POM <sup>14</sup>C models to illustrate the effort required to develop appropriate models, and the complexities involved in doing so. A robust model based on parameter values obtained from local data will be necessary to create relevant and appropriate models for use in specific countries and regions.



## Section 5: Intake & Inhalation Rates

Nuclear power plant operators calculate external and internal doses that a member of the public receives due to the operation of the nuclear power plant. People receive internal dose from carbon-14 from the inhalation and ingestion pathways. The amounts of carbon-14 that people inhale or ingest are based on the inhalation and ingestion rates. This chapter discusses these two rates and how they impact carbon-14 dose calculations.

### 5.1 Vegetation Intake Factors

Carbon-14 that is released through the atmospheric pathway is incorporated into vegetation through photosynthesis. The dose pathway of carbon-14 in vegetation to humans is through the human ingestion of produce, milk, meat, and leafy vegetables. Carbon-14 is present in milk and meat due to animals' ingestion of plant based feedstock that contains carbon-14. The dose to an individual of the public is calculated with information about the annual intake (usage factors) of various foods and the concentration of power plant related radionuclides in that food material. If site specific usage factors (based on site specific land use census) are not available, generic usage factors for individuals in different age groups can be used.

Vegetation intake (usage) factors describe the annual intake (ingestion) of produce, milk, meat, and leafy vegetables for individuals in each age group (See Equation 5-1, variable U.) Table 5-1 lists intake (usage) factors for both the average individual and a maximally exposed individual as provided in Regulatory Guide 1.109 (US NRC, 1977). Table 5-2 provides comparable parameter values from a report by Limer (2010) for the United Kingdom's (UK) Nuclear Decommissioning Authority (NDA), used for the Radioactive Waste Management Directorate's (RWMD) survey of geological repository options for the UK.

Regulatory Guide 1.109 provides for the calculation of annual organ dose from ingestion using (Equation C-13 in Regulatory Guide 1.109), Annual Organ Dose from Ingestion of Atmospherically Released Radionuclides in Food:

$$D_{ja}^D(r, \theta) = \sum_i DFI_{ija} [U_a^V f_g C_i^V(r, \theta) + U_a^m C_i^m(r, \theta) + U_a^F C_i^F(r, \theta) + U_a^L f_\ell C_i^L(r, \theta)]$$

Eq. 5-1

Where:

$C_i^V(r, \theta)$ ,  $C_i^m(r, \theta)$ ,  $C_i^F(r, \theta)$ ,  $C_i^L(r, \theta)$ : Concentrations of radionuclide, I, in produce (non-leafy-vegetables, fruits, and grains), milk, leafy vegetables, and meat, respectively, at location  $(r, \theta)$ , in pCi/kg or pCi/ℓ

$D_{ja}^D(r, \theta)$ : Annual dose to organ, j, of individual in age group, a, from ingestion of produce, milk leafy vegetable, and meat at location  $(r, \theta)$ , in mrem/yr

$DFI_{ija}$ : Ingestion dose factor for radionuclide, i, organ, j, and age group a in mrem/pCi

$f_g$ ,  $f_\ell$  : Respective fractions of the ingestion rates of produce and leafy vegetables that are produced in the garden of interest

$U_a^V$ ,  $U_a^m$ ,  $U_a^F$ ,  $U_a^L$ : Annual intake (usage) of produce, milk, meat, and leafy vegetables, respectively, for individuals in the age group a, in kg/yr or i/yr (equivalent to Uap).

Table 5-1

Usage factor values (Uap) from RG 1.109 Tables E-4 and E-5. US NRC

References: (Fletcher & Dotson, 1971; Bustad & Terry, 1956; Miller & Nash, 1971; USAEC, 1973; USAEC, 1974)

Parameter	Infant	Child	Teen	Adult
<b>AVERAGE INDIVIDUAL</b>				
Fruits, vegetables, grain (kg yr <sup>-1</sup> )	–	200	240	190
Milk (L yr <sup>-1</sup> )	–	170	200	110
Meat, poultry (kg yr <sup>-1</sup> )	–	37	59	95
Fish (kg yr <sup>-1</sup> )	–	2.2	5.2	6.9
Seafood (kg yr <sup>-1</sup> )	–	0.33	0.75	1.0
Drinking water (L yr <sup>-1</sup> )	–	260	260	370
<b>MAXIMALLY EXPOSED INDIVIDUAL</b>				
Fruits, vegetables, grain (kg yr <sup>-1</sup> )	–	520	630	520
Leafy vegetables (kg yr <sup>-1</sup> )	–	26	42	64
Milk (L yr <sup>-1</sup> )	330	330	400	310
Meat, poultry (kg yr <sup>-1</sup> )	–	41	65	110
Fish, fresh or salt (kg yr <sup>-1</sup> )	–	6.9	16	21
Other Seafood (kg yr <sup>-1</sup> )	–	1.7	3.8	5
Drinking water (L yr <sup>-1</sup> )	330	510	510	730



Table 5-2  
Human Ingestion Rates (Limer et al., 2010)

Parameter	Infant	Child	Adult
Soil Ingestion Rate (kg yr <sup>-1</sup> dry weight)	0.037	0.011	0.0037
Water Ingestion Rate (L yr <sup>-1</sup> )	260	350	600
Plant Ingestion Rate (kg yr <sup>-1</sup> fresh weight)	44	110	160
Terrestrial Animal Product Ingestion Rate (kg yr <sup>-1</sup> fresh weight)	150	170	170

Provided below is a summary of data from the United States Department of Agriculture (USDA) clearly illustrating the increases in food consumption patterns by Americans (USDA, 2002). The United States Department of Agriculture (USDA) “Agriculture Fact Book 2001-2002” details American food consumption patterns from the 1950’s through the year 2000 (See Table 5-3 below). There is a clear increase in average food consumption over the last five decades, with an increase in average daily calorie intake of 24.5% (USDA, 2002). Average yearly meat consumption has increased by 26 kilograms since the 1950’s, from 63 kg per year to 89 kg per year. Per capita consumption of fruit and vegetables increased by 20% from the 1970’s to the year 2000, from 267 kg total fruit and vegetables per year to 322 kg. Grain consumption increased by 45% from 1970-2000, from 63 kg total grain products to 91 kg (USDA, 2002). More data can be found in the following references, but the numbers provided in tables represent the most recent available data (McKone, 1989; McKone, 1994; Rupp, 1980; Rupp et al., 1980; Yang & Nelson, 1986).

There are two primary differences in the above tables. The first is the difference between the maximally exposed individual values that NRC uses and average consumption rates (Table 5-1). As the data shows, usage factor values for the maximally exposed individual are significantly higher than average. While the maximally exposed individual approach is indeed the most conservative method possible, it is also relatively unrealistic of actual consumption rates. As the data from Limer et al. 2010 shows, parameter values for plant consumption rates are less than the averages from the RG 1.109 data. For plant ingestion rates, RG 1.109 uses a value of 200 kg/year for children, while Limer et al. 2010 uses a value of 110 kg/year. Similarly, for plant ingestion rates for adults, RG 1.109 uses a value of 190 kg/year while Limer et al. 2010 uses a value of 160 kg/year. Secondly, vegetation consumption rates for infants are not addressed in RG 1.109, but a value is given for infants in the Limer et al. 2010 data (Table 5-2). The comparisons suggest that a closer look at average yearly consumption rates is necessary in order to obtain realistic data.

Table 5-3  
USDA Data on Human Consumption Rates (USDA, 2002)

Parameter	1950-59	1960-69	1970-79	1980-89	1990-99	2000
<b>Total Meats (kilogram per year, kg yr<sup>-1</sup>)</b>	62.8	73.5	80.5	82.8	85.9	88.7
Red Meats (kg yr <sup>-1</sup> )	48.5	55.6	58.8	55.4	51.1	51.6
Poultry (kg yr <sup>-1</sup> )	9.3	13.0	16.0	21.0	28.1	30.2
Fish/Shellfish (kg yr <sup>-1</sup> )	5.0	4.9	5.7	6.5	6.7	6.9
<b>Eggs (number)</b>	374	320	285	257	236	250
<b>Total Fruits and Vegetables (kg yr<sup>-1</sup> fresh weight)</b>	–	–	267.0	282.8	312.9	321.7
Fresh Fruit (kg yr <sup>-1</sup> fresh weight)	–	–	45.2	51.4	56.2	57.6
Fresh Vegetables (kg yr <sup>-1</sup> fresh weight)	–	–	67.2	71.5	82.7	91.7
<b>Total Grain Products (kg yr<sup>-1</sup>)</b>	70.6	64.8	62.8	71.5	86.6	90.9
Corn Products (kg yr <sup>-1</sup> )	7.0	6.3	5.0	7.9	11.1	12.9
Rice (kg yr <sup>-1</sup> )	2.4	3.2	3.3	5.1	8.0	9.0

## 5.2 Inhalation Rates

Regulatory Guide 1.109 provides for the calculation of annual organ dose from inhalation using Equation 5-2.

$$D_{ja}^A(r, \theta) = R_a \sum_i \chi_i(r, \theta) DFA_{ija} \quad \text{Eq. 5-2}$$

(note that this is equation C-4 in Regulatory Guide 1.109 (USNRC, 1977b))

Where:

$D_{ja}^A(r, \theta)$  is the annual dose to organ j of an individual in age group a at location  $(r, \theta)$  due to inhalation, in mrem/yr

$DFA_{ija}$  is the inhalation dose factor for radionuclide i, organ j, and age group a.

$R_a$  is the annual air intake for individuals in age group a, in m<sup>3</sup>/yr

$\chi_i(r, \theta)$  is the annual average concentration of radionuclide  $i$  in air at location  $(r, \theta)$ , in pCi/m<sup>3</sup>

Although the chemical form of <sup>14</sup>C is an important consideration in the adsorption of <sup>14</sup>C in the form of CO<sub>2</sub> by vegetation. The chemical form of <sup>14</sup>C is not mentioned as a consideration for inhalation dose in the regulatory guidance provided in Regulatory Guide 1.109 (US NRC, 1977b). Additional information can be located in Section 3.1 and Appendix A of “Carbon-14 Dose Calculation Methods at Nuclear Power Plants.” (EPRI, 2012).

A review of the inhalation rates from Regulatory Guide 1.109 and ICRP Publication 66 “Human Respiratory Tract Model for Radiological Protection” (ICRP, 1994) are displayed in Table 5-4 and Table 5-5. Although there are variations in the age groups used in these two references and various states of activity in ICRP 66, the values provided by Regulatory Guide 1.109 appear generally consistent with ICRP 66, given this, it is expected that there will be little impact on dose calculations from variations in inhalation rates (annual air intake).


*Table 5-4*  
*Breathing rates from RG 1.109 (US NRC, 1977)*

Group	Inhalation Rates (m <sup>3</sup> yr <sup>-1</sup> )			
	Infant	Child	Teen	Adult
Average Individual	–	3700	8000	8000
Maximally Exposed Individual	1400	3700	8000	8000

*Table 5-5*  
*Breathing rates from ICRP 66 (ICRP, 1994)*

Group	Inhalation Rates (m <sup>3</sup> yr <sup>-1</sup> )			
	Sleeping	Sitting	Light Exercise	Heavy Exercise
Adult Male	3945	4734	13149	26298
Adult Female	2805	3419	10958	23668
15 y old Male	3682	4208	12097	25597
15 y old Female	3068	3506	11396	22529
Child 10 y old	2717	3331	9818	17795
Child 5 y old	2104	2805	4997	–
Infant 1 y old	1315	1929	3068	–
Infant 3 month old	789	–	1666	–





## Section 6: Enhancements to Carbon-14 Dose Calculation Methodologies

The purpose of this project is to explore the latest scientific knowledge related to carbon and carbon-14 ( $^{14}\text{C}$ ) science to identify opportunities for and provide technical bases for enhancing the accuracy of carbon-14 dose calculations. This report addresses, in some detail, the presence and the various sources of  $^{14}\text{C}$  in the environment as well as provides recent technical information and models that would be of value to the computation of the dose from  $^{14}\text{C}$  in the effluents of nuclear generating stations.

This section discusses, in summary level, the key findings of the research with specific emphasis on findings that impact or enhance the accuracy of carbon-14 dose calculation methodologies. These findings can be used by nuclear power plant operators to calculate more accurate, site-specific doses to the public due to carbon-14. These findings may also inform regulatory bodies when they revise applicable regulatory guidance.

The United States Nuclear Regulatory Commission (US NRC) intends to update radiation protection related regulations and regulatory guides to reflect updated dose assessment science and terminology. (US NRC, 2012) Regulatory Guide 1.109, Revision 1 “Calculation of Annual Doses to Man from Routine Releases of Reactor Effluents for the Purpose of Evaluating Compliance with 10 CFR Part 50, Appendix I” (US NRC, 1977b) may be included in this revision process. As such, potential revisions to enhance the accuracies of carbon-14 dose assessments using Regulatory Guide 1.109 is provided in this section for consideration. Refer to Sections 4.2.1.3 and 5 for additional details on the Regulatory Guide 1.109 methodology.

### **6.1 Chemical Form of Carbon-14 in Nuclear Generating Station Effluents**

*Recommendation for Enhanced Accuracy: Use the fraction of carbon-14 in gaseous effluents that is in the form of  $\text{CO}_2$  for dose calculations.*

Carbon-14 is incorporated into the human dose pathway through the photosynthesis. Although 1-2% of plant carbon is absorbed through the roots, it should be noted that surface soil concentrations of  $^{14}\text{C}$  will be in equilibrium with atmospheric levels of  $^{14}\text{CO}_2$ . In excess of 98% of the plant carbon is atmospheric  $\text{CO}_2$  incorporated during photosynthesis. Furthermore, the high percentage of  $^{14}\text{C}$  absorbed by vegetation from airborne sources supports the use of the specific activity models presented in Section 4.2.1.3 from Regulatory Guide 1.109.

Due to the reasons discussed above and in Section 4, the fraction of  $\text{CO}_2$  in nuclear power plant effluents is a critical consideration in calculating the dose resulting from  $^{14}\text{C}$  in the effluents of nuclear power plants. Many nuclear generating stations are currently using the fraction of carbon-14 that is in the form of  $\text{CO}_2$  in their dose calculations, since only the  $^{14}\text{CO}_2$  is subject to incorporation into vegetation. This practice leads to more accurate calculation of carbon-14 dose due to nuclear power plant gaseous effluents and is documented as a good practice in EPRI Report 1024827 "Carbon-14 Dose Calculation Methods at Nuclear Power Plants" (EPRI, 2012).

The relative production of  $^{14}\text{C}$  in the chemical form of  $\text{CO}_2$  and hydrocarbons varies by reactor type. Table 6-1 contains data taken from "Management of Waste Containing Tritium and Carbon-14 published by the International Atomic Energy Agency" (IAEA) (Technical Report Series No. 421; IAEA, 2004), illustrates this point.

Research documented in EPRI Report 1021106 "Estimation of Carbon-14 in Nuclear Power Plant Gaseous Effluents" (EPRI, 2010) and data collected by the United States Nuclear Industry presented in EPRI Report 1024827 "Carbon-14 Dose Calculation Methods at Nuclear Power Plants" (EPRI, 2012) is also consistent with the above data in Table 6-1. However, it is important to note that presence of a recombiner in a PWR gaseous effluent systems will result in the release of mostly  $^{14}\text{CO}_2$  rather than organic (or hydrocarbons) forms of  $^{14}\text{C}$  (EPRI, 2010).

*Table 6-1  
Comparisons of Chemical Form of Carbon-14 in Airborne Release by Reactor Type  
(IAEA, 2004)*

Reactor Type	$^{14}\text{CO}_2$	$^{14}\text{C}$ hydrocarbons
Pressurized Water Reactor (PWR) (USA and Europe)	5-25%	75-95% $\text{CH}_4$ and $\text{C}_2\text{H}_6$
Boiling Water Reactor (BWR) (USA and Europe)	80-95%	5-20%

Regulatory Guide 1.109 assumes that all  $^{14}\text{C}$  is in the chemical form of “in the oxide form ( $\text{CO}$  or  $\text{CO}_2$ )”. (NRC, 1977b). Including a factor in this method for the percentage of  $^{14}\text{CO}_2$  using a conservative value based on the reactor type or studies that establish the percentage of  $^{14}\text{CO}_2$  of the total  $^{14}\text{C}$  in the airborne effluent would increase the accuracy of the resulting dose calculation.

## 6.2 Total Plant Mass that is Natural Carbon

*Recommendation for Enhanced Accuracy: Evaluate or consider up-to-date and plant specific values for total plant mass that is natural carbon.*

The total plant mass that is natural carbon varies by plant type. The IAEA, in the “Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments,” (IAEA, 2012) provides values for individual type of plant and animal products<sup>15</sup>.

These values can be found in Table 4-2 and Table 4-3 of this report. The U.S. NRC assigns a single value of 0.11 to total plant mass that is carbon in Regulatory Guide 1.109 (See Equation 4-3).

Table 6-2

Comparison of Mean Fraction of Carbon/Total Plant Mass IAEA Handbook 472 Table 66 (IAEA, 2012) with Regulatory Guide 1.109 Value of 0.11

Plant Category	Mean Fraction of Carbon/Total Plant Mass	% Ratio of .11 to the Mean Fraction of Carbon Equation 4-3
Grass (Fodder)	0.010	1100%
Leafy & Non Leafy Vegetables	0.030	367%
Root Crops	0.046	239.1%
Fruit	0.062	177.4%
Tubers	0.103	106.8%
Maize(Sweet Corn)	0.120	91.7%
Silage	0.130	84.6%
Maize (Feed Corn)	0.380	28.9%
Cereals	0.39	28.2%
Leguminous Vegetables (Seed)	0.41	26.8%
Leguminous Vegetables (Vegative Mass)	0.59	18.6%

<sup>15</sup> Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments (IAEA, 2012) provides 272 references, many of which are based on actual measurements.

The selection of 0.11 for the total plant mass that is carbon may be conservative or non-conservative depending of the vegetation selected. This is demonstrated in Table 6-2. Vegetation such as seeds, cereals, feed corn, sweet corn, would be non-conservative (estimating a lower concentration of carbon-14 and a lower dose). Leafy and non leafy vegetables, root crops, tubers, and fruit would be conservative (overestimating the concentration of carbon-14 and higher doses).

The use of plant specific total plant mass carbon, such as those provided in IAEA Handbook 472, would support more accurate, site specific calculation of carbon-14 doses.

The accuracy of the methodology provided in Regulatory Guide 1.109 would be enhanced if the US NRC staff considers providing clarification on the use of 0.11 for the total plant mass that is carbon, and allowing the use of values from IAEA Handbook 472.

### **6.3 Concentration of Natural Carbon in the Atmosphere**

*Recommendation for Enhanced Accuracy: Use up to date value for concentration of natural CO<sub>2</sub> in the atmosphere.*

Throughout the past few decades, the concentration of natural carbon in the atmosphere has increased. In Regulatory Guide 1.109, published in 1977, the US NRC staff establishes the fraction of carbon in the atmosphere as 0.16 gC/m<sup>3</sup>. (US NRC. 1977b) The IAEA more recently established the concentration of natural carbon in the air as 0.20 gC/m<sup>3</sup> in the “Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environment” (IAEA, 2010). The value of 0.16 gC/m<sup>3</sup> used by the NRC staff (US NRC. 1977b) would lead to a 25% over-estimation of the concentration in vegetation using Equation 4-3.

Based on the evidence provided by current scientific knowledge, the use of a value reflecting the current atmospheric concentrations of CO<sub>2</sub> would lead to more accurate calculation of carbon-14 doses.

The accuracy of the methodology provided in Regulatory Guide 1.109 would be enhanced if, assuming that Equation 4-3 is retained, the value of 0.16gC/m<sup>3</sup> is revised to a value reflecting the current atmospheric concentrations of CO<sub>2</sub>. Additionally, given that the concentration of CO<sub>2</sub> in the atmosphere continues to increase, providing a mechanism for updating the atmospheric concentration of CO<sub>2</sub> would ensure that carbon-14 dose estimates continue to remain accurate over time (see Section 4.3.1 of this report for a detailed discussion).

### **6.4 P-Factor or Fractional Equilibrium Ratio**

The p-factor is found in US NRC Regulatory Guide 1.109 (See Equation 4-3). It is identified as the parameter (p) and described as the fractional equilibrium ratio as follows:



In the case of intermittent releases such as from gaseous waste decay tanks the parameter  $p$  is employed to account for the fractional equilibrium ratio. The parameter  $p$  is defined as the ratio of the total annual release time (for C-14 atmospheric releases) to the total annual time during which photosynthesis occurs (taken to be 4400 hours). Under this condition the value of  $p$  should never exceed unity. For continuous C-14 releases,  $p$  is taken to be unity. (US NRC, 1977b)

No references for the use of the P-factor were noted in the literature review preformed for this report except Regulatory Guide 1.109 (US NRC, 1977b).

## 6.5 Carbon Incorporation During Photosynthesis

*Recommendation for Enhanced Accuracy: Evaluate or consider using meteorological dispersion from daylight growing season hours in computation of carbon-14 doses.*

Since  $^{14}\text{C}$  is primarily incorporated into vegetation through photosynthesis, Killough & Rohwer recommended that the meteorological dispersion,  $\left[\frac{X}{Q}\right]$ , be “renormalized such that they reflect average ground level air concentrations,  $[X]$ , for daylight growing season hours.” (Killough & Rohwer, 1978). Additional detail regarding this recommendation is in Section 4.2.1.2 of this report.

Duke Power evaluated the implications of limiting the dose calculations to daylight hours or during the period of photosynthesis for the Catawba Nuclear Station (EPRI, 2012) (Duke Energy, 2011a and 2011b). They found that meteorological dispersion,  $\left[\frac{X}{Q}\right]$ , that would be used for the carbon-14 dose calculations decreased in most sectors, but not all sectors using this approach. This assessment was not used for the computation of dose in the Duke Power 2010 Annual Radioactive Effluent Release Report (ARERR). Additional detail on how the calculations may be performed and more detailed results are found in Section 3.2.6 of “Carbon-14 Dose Calculation Methods at Nuclear Power Plants” (EPRI, 2012).

The use of meteorological dispersion  $\left[\frac{X}{Q}\right]$  from daylight growing season hours would lead to a more accurate computation of dose as a result due to carbon fixation in vegetation only during the period of photosynthesis and the growing season. Including a consideration of this in any future revision of Regulatory Guide 1.109 would enhance the accuracy of carbon-14 dose calculations using that methodology.

Note that it is important, when performing dose calculation using periods of analysis other than a calendar year, that the  $\left[\frac{X}{Q}\right]$  and the source term  $[Q_{14}]$  are for the same periods (i.e., daylight hours and/or growing season). Daylight meteorology is inherently more unstable than nighttime meteorology, resulting in greater meteorological dispersion.

## 6.6 Consumption Rates of Vegetation

*Recommendation for Enhanced Accuracy: Conduct a complete literature review and update food consumption rates.*

Data from the United States (US) Department of Agriculture show changing patterns in the consumption of various food stuffs (USDA, 2002). A study conducted by Limer (2010) for the United Kingdom (UK) also provides additional data related to consumption rates, including vegetation consumption rates for infants. (Limer, 2010) See Section 5.1 for additional detail (Equation 5-1 and factors  $U_a^V$ ,  $U_a^m$ ,  $U_a^F$ , and  $U_a^L$ ). These more recent studies suggest that the mean consumption rates are less than those provided in Regulatory Guide 1.109 for some age groups.

The availability of updated literature and data related to consumption rates indicates that a more complete literature review should be undertaken to update the consumption rates of various food stuffs that are provided in Regulatory Guide 1.109 should a revision of the guidance be undertaken.

## 6.7 Dose Factors

*Recommendation for Enhanced Accuracy: Incorporate up-to-date dose commitment factors into the calculation of doses from nuclear generation station effluents.*

Since Regulatory Guide 1.109 (US NRC, 1977b) was published 35 years ago, science and research has yielded updated dose models as published in ICRP 103 (ICRP, 2007). The current guidance is now more than 35 years old and may be revised in a normal review cycle at NRC or as part of the proposed implementation of ICRP 103. See Section 2 for further detail regarding dose factors. The NRC dose factors based on ICRP 103 were not yet available at the time this report was written. A robust comparison of Regulatory Guide 1.109 carbon-14 dose factors with those of ICRP-72 is contained in EPRI Report 1024827 “Carbon-14 Dose Calculation Methods at Nuclear Power Plants” (EPRI, 2012).

The US NRC intends to update radiation protection related regulations and regulatory guides to “align with the most recent methodology and terminology for dose assessment.” (US NRC, 2012) Should the NRC staff revise Regulatory Guide 1.109 as part of this process, the dose factors used in Regulatory Guide 1.109 should be updated to more recent guidance such as ICRP Publication 103 “Recommendations of the International Commission on Radiological Protection” (ICRP, 2007). Such a revision would be based on all the isotopes of interest to nuclear power plants, not just  $^{14}\text{C}$ .

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# Appendix A: Radiocarbon Laboratory Sample Collection, Handling, and Shipping

Radiocarbon laboratories use very sensitive analytical processes specifically designed for environmental levels of  $^{14}\text{C}$ . Samples for a radiocarbon laboratory can pose serious contamination issues if they are significantly higher than environmental levels of  $^{14}\text{C}$ -14. This appendix was provided courtesy of Beta Analytic and provides general guidance in the collection, shipping, and handling of samples for radiocarbon analysis.

## General Cautions:

- *Never* ship samples of plant effluents to a radiocarbon laboratory
- Only ship plant and animal parts or products
- Do not ship surface water, soils, or sediments
- Never handle or store nuclear plant samples in areas with samples for radiocarbon analysis, nor ship samples from radiological effluent laboratories
- Always discuss your samples with the radiocarbon laboratory prior to sending

Important to remember is that upon “death”, respiration ceases and the  $^{14}\text{C}$  signature of the material is defined. “Death” could be when a leaf falls off a branch, when a berry drops to the ground or when the corn or hay is harvested. As such, the vegetation does not have to be living at the time of collection to obtain environmentally relevant data. Corn stalks or sugar cane from the last harvest will show NPP  $^{14}\text{CO}_2$  uptake just as well as freshly collected corn or sugar cane. The only difference will be when the last time  $^{14}\text{C}$  was incorporated into their cellular structures.

Vegetation is the most appropriate material for analysis. A single piece of fruit or vegetable or a few grains of wheat or rice are enough for an analysis. Collecting samples with bare hands is fine but wearing clean gloves is a good practice. Collection, storage and shipping in plastic bags is acceptable. Samples subject to rot or biological decay, such as fruits and vegetables should be stored in a refrigerator until shipping and then shipped by overnight courier.

The life cycle of the material collected and analyzed will determine the length of time represented in the result (e.g., longer life cycle = longer average represented by the  $^{14}\text{C}$  signature). The most recent  $^{14}\text{C}$  uptake will be seen in living green plant parts which are in direct contact with the air (e.g., corn husks, wheat blades, hay, leaves, grass blades) whereas materials such as beans, berries, seeds, nuts, and corn kernels will represent a longer period of uptake.

It is highly recommended that your indicator stations be chosen to be realistic indications of the environmental impact from the station effluents as described in the ODCM or ODAM for the station. Control stations are highly recommended to be from areas not impacted by the station effluents.

Best consistency in results will come from like-materials between control vs. indicator samples. For example, corn kernel (non-impacted) vs. corn kernel (impacted) or wheat grain (non-impacted) vs. wheat grain (impacted) is good practice.

A minimum of 3 samples is recommended from both the control location and each of the indicator location sites ( $^{14}\text{C}$  results to be averaged for dose calculations).

## **A.1 Summary: Guidelines for Sample Collection, Handling, and Shipping**

### **A.1.1 Material for Analysis**

- Ideally select vegetation that has green leafy parts in direct contact with air.
- It is fine to collect remnants of harvested crops if it is the only material available.
- Avoid anything rotten or moldy.

### **A.1.2 Quantity Required**

- A single piece of fruit or vegetable.
- A few grains of wheat or rice.

### **A.1.3 Sample Handling and Labeling**

- You do not need to wear gloves when collecting samples (but it is good practice to do so).
- Refrigerate food crops if not shipping immediately
- Collection and storage in zip-lock bags is acceptable.
- Decide on your sample labeling codes prior to going into the field.
- Label your samples in the field at the time of collection. Don't wait until later when it may get confusing. Take a Sharpie or other indelible ink pen with you.
- Fill out any submittal forms required by the laboratory.

- It is fine to dry the samples, but you do not need to do so.
- Ship by courier with tracking options (in case the samples do not arrive at the laboratory when expected).
- Provide the approximate date of “death” to the laboratory. This will be when you picked the corn cob from the stalk or when it was harvested.
- Tell the laboratory what units you wish the results to be reported in (Bq/kg mass, pCi/kg mass).







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Low-Level Waste and Radiation Management

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