Master of Science Thesis

OCCUPATIONAL EXPOSURE of ^{14}C: a systematic investigation of ^{14}C contamination of workers at the nuclear power industry, the pharmaceutical industry and other laboratories using ^{14}C.

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Contents

Abstract

With the help from accelerator mass spectrometry (AMS), hair from workers within the nuclear power industry, pharmaceutical and biotechnology industry has been investigated to see if contamination from ^{14}C. ^{14}C-using companies and nuclear power plants were contacted, volunteers were recruited, who sent in hair samples together with a questionnaire. The samples were washed and prepared for ^{14}C analysis with a single stage AMS (SSAMS) machine. Different ways were tried to remove sulphur impurities form the hair samples. Hair collected at hairdresser saloons was analysed to act as a background indicator. Contaminations were found in two samples that were in the order of 12 and 78 times higher than normal background. Also smaller contaminations were found in 12 samples that indicate a general contamination in the work environment. Further studies are needed to investigate if the contamination is entirely internal, and not to some extent external ^{14}C absorbed by the hair. If the ^{14}C contamination is mainly internal, this work suggests that the workers at least should be made aware of the amount of ^{14}C they receive internally. The work presented in this master's thesis shows that our graphitization method is good for hair and AMS can be used as a tool to monitor small amounts of ^{14}C in hair.

Acronyms

SSAMS	Single Stage Accelerator Mass Spectrometry
MS	Mass Spectrometry
BP	Before Present (Present being defined as the year 1950 AD)
NBS	United States National Bureau of Standards
Oxalic Acid	$H_2C_2O_4$. A poisonous strong acid that occurs in various plants as oxalates and is used especially as a bleaching or cleaning agent and as a chemical intermediate
LMFBR	Liquid Metal Fast Breeder Reactor
pMC	Percent Modern Carbon, 1pMC=2.26Bq/kg kol

1. Introduction

In this master's thesis, a study conducted with accelerator mass spectrometry (AMS), is made for investigation of the amount of ^{14}C in workers of the nuclear power industry, the pharmaceutical industry and other laboratories. The ^{14}C concentration in hair collected from workers potentially exposed to the radio nuclide, is used as a rough measure of the time accumulated exposure from ^{14}C that the person has been exposed to. As a pure β-emitter with a maximum energy of 156 keV, ^{14}C is not usually monitored in workers since the detection of the radionuclide requires special chemical preparation and detection techniques. It is therefore of great interest to gather information on the extent of the occupational exposure to workers. In this work, optimisation of the graphitisation process had to be made. Much effort was devoted on choosing the right combination of water, Ag, Cu and H_2O to absorb the sulphur from the hair samples.

There are very few reports concerning the ^{14}C contamination of radiation workers in the literature. In one such report [1], a university medical centre was found to be highly contaminated throughout by tritium- and ^{14}C-containing compounds. Incidents or accidents involving ^{14}C are somewhat more frequently described in the literature. One such article describes an incidence of high-level contamination in a laboratory producing ^{14}C-labelled barium carbonate [2]. An estimated committed effective dose of 3 mSv was reported for one member of staff.

Although data concerning the ^{14}C contamination of radiation workers are scarce, reports on ^{14}C contamination in the environment around nuclear reactors and laboratories using ^{14}C are more frequently found in the literature (see review by Stenström et al [3]). A rather extreme case has been reported by Leprieur et al [4]. Contamination by ^{14}C inside and around a company specializing in radioactive organic molecule synthesis was investigated. Inside the laboratory, the surface contamination reached values above 20 000 Bq/cm^2. Within a 50 m radius of the facility, 66 000 Bq/kg C was found in soils, and 25 000 Bq/kg C in plants. If the latter value is assumed to be the static specific activity in humans, this would result in an effective dose rate of 1 mSv/year.

AMS is a technology not only used for radiocarbon dating, but also used in such diverse applications such as nutritional, environmental, medical and occupational studies. Biological samples are also studied with AMS to calculate when cells in various human organs were formed. This is made possible with a relatively new method called bomb pulse dating. Bomb pulse dating utilises the increase in ^{14}C from nuclear testing in the 1950s and 60s, to estimate the age of cells. Bomb-pulse dating can be used in a variety of applications such as drug dating, wine authentication, forensic medicine and much more [5, 6, 7, 8, 9].

2. ^{14}C: background and applications

2.1 Health Effects of 14C

^{14}C is one of three naturally occurring carbon isotopes (10^{-10}% ^{14}C, 1.1%^{13}C and 98.9% ^{12}C) and is produced both cosmogenically in the upper atmosphere and in for example nuclear facilities and by nuclear detonations.^{14}C is a beta emitter with a maximum energy of 156.48 keV and a mean energy of 49 keV. The physical half-life is 5730 years, and the biological half-time has been estimated to be 40 days. However, the effective half-life may be longer/shorter depending on what it is bound to. The estimated effective half-life of 40 days is

thus only applicable on ^{14}C that has been widely distributed throughout the environment and basically produced a uniform contamination of the diet [10]. Natural ^{14}C accounts for about 12 μSv/year [11]. In the nuclear power industry ^{14}C appears in airborne effluents mainly bound to $^{14}CO_2$ and $^{14}CH_4$ depending on which reactor type [10]. Thus, workers in the nuclear power industry are potentially exposed to inhalation of these substances. When it comes to the pharmaceutical, biotechnology and medical industry there are also other substances that ^{14}C are bound to, such as sugar, fatty acids, medicine etc.

From a radiation protection point of view, the risk of external exposure is negligible, e.g. if ^{14}C is placed on a table in front of a person, the risk of acquiring an exposure of significance is small, due to the low beta energy of ^{14}C. However, there may be a risk of acquiring doses of importance if ^{14}C is ingested, because it behaves as natural carbon. ^{14}C can also enter the human body through inhalation or dermal exposure. Since there are many different compounds in the pharmaceutical field and other fields some reason that there should be an ALI for these compounds also, but according to [10] there is little evidence to support that view.

2.2 Hair as a bio-indicator of the amount of ^{14}C

In this master's thesis, hair is used as an indicator of the accumulated exposure under a certain time. Hair is used as an indicator of bio-exposure in a variety of fields, such as heavy metals, medical studies, medical diagnosis in hospitals, although some correlations between body content and hair concentration is questioned such as in nutritional studies. For example some essential minerals may show in hair when there is an excess of the substance but not a deficiency. A more accepted type of study is the investigation of heavy metals in humans and animals. [12-17]

The relative ^{14}C content in hair have been used in forensic medicine to determine the time of death for murder victims, using so called Bomb Peak Dating. Nuclear bomb tests in the 50s and 60s lead to an in increased amount of ^{14}C in atmospheric carbon dioxide. After the nuclear test ban treaty in 1963 the amount of ^{14}C in the atmosphere decreased rapidly due to the large uptake in oceans and biosphere. These changes in the amount of ^{14}C are called "bomb peak" and can be used as a calibration curve for very young samples. To obtain a reliable measure of when the victim deceased a sample with a rapid turnover time of carbon is needed, e.g. hair. Research has suggested that the year the victim deceased can be accurately estimated from ^{14}C analysis from the deceased hair. [5,6]

In this study a background of ^{14}C is measured in unexposed hairs, and subtracted from the amount in the test subject's hair. This is to see if there is any excess of ^{14}C in the hair and so also in the body.

2.3 Measuring techniques

There are a few different techniques to determine ^{14}C concentration in various samples, for example liquid scintillation counting and proportional counting. Proportional counting consists of a gas filled volume with two electrodes. The ion pairs created by the ionizing radiation are measured and then related to the number of decays and energy. In liquid

scintillation counting the sample is put in a solvent that produces photons when interacting with the beta particles. The vials are put in a light tight box with PM tubes where the scintillation light is measured. These two techniques have relatively poor efficiency compared to AMS. These techniques are also time consuming and requires a larger amount of the sample, because ^{14}C has so long half-life. It may also require additional sample preparation to separate ^{14}C from other nuclides.

AMS first came to use in low-energy nuclear physics laboratories in the late 1970s. It was needed for radiocarbon dating of small samples (<<1 g). The first AMS-facilities were thus mostly used for ^{14}C-dating, although today AMS is used in climate research, biomedical and pharmaceutical research, forensic medicine etc, combined with several different nuclides. [18].

AMS is an extension of mass spectrometry (MS), a traditional method for determining the masses of specific positive or negative ions. In MS, the sample is introduced into an ion source, which produces a positive or negative ion beam from the sample. The ions emerging from the ion source are generally accelerated in vacuum to energies in the keV range by a strong electric field. In the most basic MS system, a magnetic field applied perpendicular to the ion beam will bend off different ions from the initial ion trajectory according to their mass to charge ratio (m/Z). A detector is used to register just one specific m/Z ratio at a time. By varying the magnetic field, a spectrum of various m/Z ratios is obtained, yielding information e.g. about the abundance of various ions, or isotopes, in the sample. MS can however not be used for measurements of ^{14}C, because of an insufficient detection limit, and because of interfering atomic or molecular isobars, e.g. $^{14}N^+$ if using positive ions and $^{13}CH^-$ if using negative ions.

AMS has many advantages over regular mass spectrometry, such as the ion source that produces singly charged negative ions, which reduces the background from such nuclides as ^{14}N. This is because ^{14}N does not form stable negative ions. No other element comprise of naturally occurring mass 14 isotopes (all are radioactive with extremely short half-lives). Thus, if using a negative ion source, the interference will only be negatively charged molecular isobars. The technique of AMS has the ability to prevent these interfering molecular isobars from reaching the detector of the AMS system, as described below.

The AMS-facility in Lund is a Single Stage Accelerator Mass Spectrometry (SSAMS) system produced by National Electrostatics Corporation (NEC), Middleton, Wisconsin, USA, see figure 1.

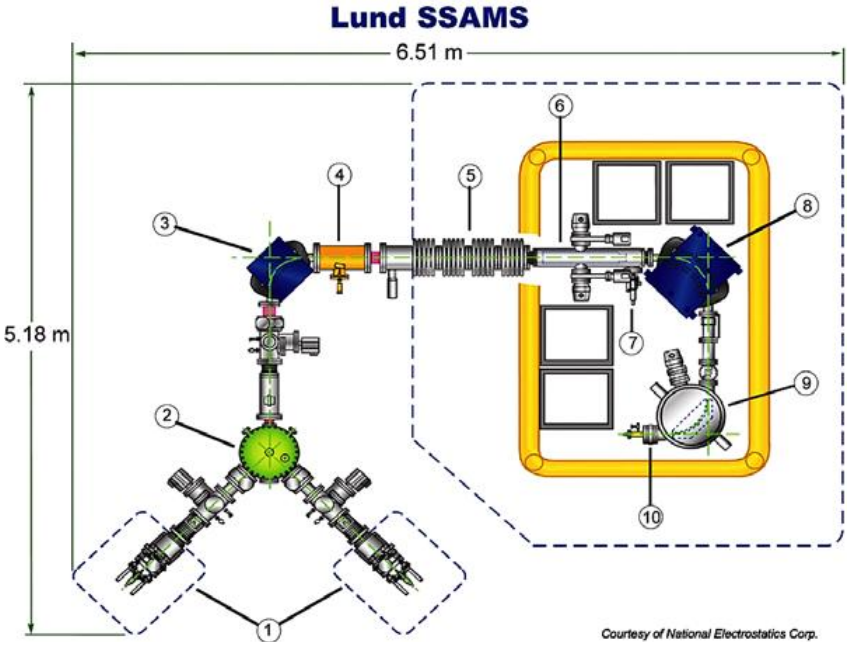

Fig. 1. The Lund SSAMS facility. 1.) Ion sources; 2.) Rotable spherical electrostatic analyzer (ESA); 3.) Dipole magnet; 4.) Einzel lens; 5.) 250kV accelerator; 6.) Molecular dissociator; 7.) Argon Valve; 8.) Dipole Magnet; 9.) Spherical electrostatic analyser (ESA); 10.) Sequential postaccelerator deflector (SPAD); The 12C and 13C Faraday Cups are placed off-axis, just after the high energy magnet.

This was installed in May 2003, and is the first low energy, open air, single stage AMS system. As seen in the figure 1, the system is equipped with two ion sources, which conveys that e.g. one can be used for low activity measurements, and one for high activity measurements, with low risk for cross-contamination [19]. The ion source used in the SSAMS system is a so-called SNICS-II, i.e. Source of Negative Ions by Caesium Sputtering.

A view of the ion source can be seen in figure 2. Caesium vapour, created in an oven, emerges into an enclosed area. Some of this caesium is ionised and thus accelerated towards the cathode where the 40-sample wheel is placed (each cathode contains elemental carbon extracted from a sample to be analysed). When the caesium ions hit the sample negative carbon ions are created, and the ion beam is given an initial energy of about 40 keV. The electrostatic analyser (ESA, no. 2 in figure 1) can easily switch between the two ion sources and improves the energy resolution of the injected beam. A first mass selection is performed using a magnetic field (the dipole magnet: no. 3 in figure 1), separating the masses 12, 13 and 14 into separate ion beams, the latter consisting mainly of $^{14}C^-$, but also of e.g. $^{13}CH^-$ and $^{12}CH_2^-$.

The separate mass-selected ion beams are sequentially accelerated over a voltage of 250 kV in the accelerator tube. After the accelerator there is a molecular dissociator, which consists of a thin gas of Ar. Here the molecules have enough energy to break apart when colliding with the Ar gas. After the molecular dissociator, the charge state of the ions has changed from -1 to on average +1. The former molecular isobars, which now have been split up into positively charged elemental fragments, are removed from the mass 14 beam by magnetic and electric fields (nos. 8 and 9 in figure 1).

The ^{14}C-ions are measured in a detector at the end of the line and $^{12}C/^{13}C$ are measured in separate Faraday cups (placed in two offset positions just after the high-energy dipole magnet: no. 8 in figure 1).

The ^{14}C specific activity of each sample is determined by correcting the measured $^{14}C/C$ ratios for background (using the blanks), and comparing the $^{14}C/C$ ratios to those of the standard samples of known activity (also corrected for background).

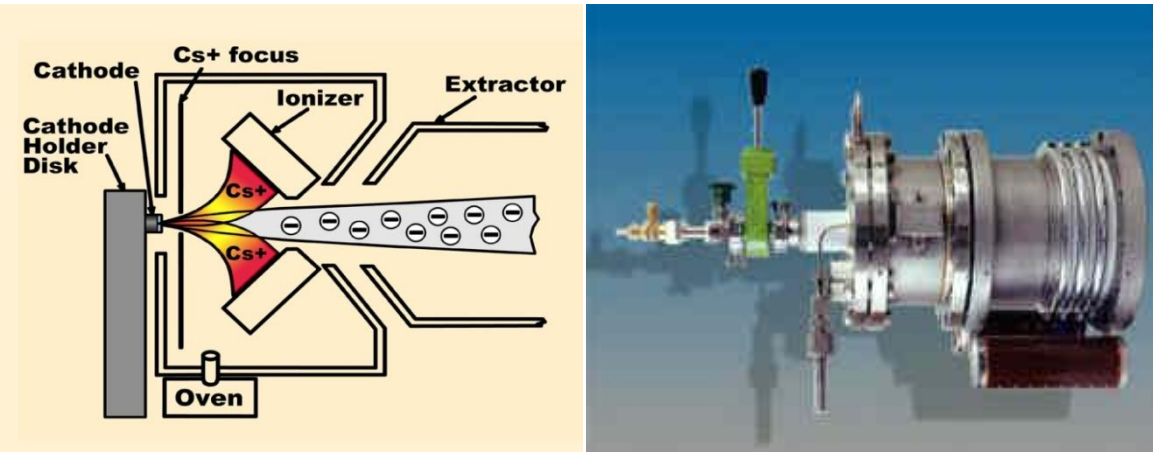

Fig. 2. Internal view of the SNICS-II ion source used in the Lund SSAMS system and external view of the ion source.

The most apparent difference in a SSAMS system from a regular AMS system is that the second acceleration step is omitted. In a regular AMS system, there is a second acceleration step after the molecular dissociation, but in SSAMS it goes directly to the analyse step. The analyzing magnet, electrostatic spherical analyzer and detector are all mounted on a 250 kV deck, which is among the lowest voltage AMS systems available. An advantage of this SSAMS machine is that it can produce comparable results of machines with higher voltage, but the SSAMS requires 40% less floor space and costs about 30% less. Every part is also accessible all the time. At the present moment the SSAMS in Lund has a resolution of ±50 years, but it is thought that the resolution will be as low as ±30 years.

2.4 Sample Preparation

Before the samples can be analyzed in the AMS-system, samples have to be prepared by a process called graphitization in which samples are converted into solid graphite. The procedure starts with combustion of the sample in presence of CuO in a vacuum system. The CO_2 produced in the combustion is cryogenically purified from other combustion gases by using two different cold traps. The first cold trap contains ethanol, cooled to a temperature just above the freezing point of CO_2, to remove water vapour and gases condensable at temperatures above the freezing point of CO_2. In the second cold trap, the CO_2 is frozen by the help of liquid nitrogen, and vacuum is pumped to remove gases below the freezing point of liquid nitrogen. The purification is necessary for good performance of the subsequent process, in which the CO_2 is reduced to elemental carbon (graphite) over a heated iron catalyst in the presence of hydrogen gas. But since hair contains sulphur it slows down the graphitization process. Therefore a modification to the process was made that is described in greater detail later.

3. Materials and Methods

3.1 Sample collection

To be able to collect the hair samples for this study, an ethics application had to be sent in and approved by the local ethical vetting board in Lund.

For collection of samples, nuclear power plants, biotech/pharmaceutical companies, research facilities etc. were localized and contacted. These companies were chosen because of their extensive ^{14}C usage and nuclear power companies for their exposure to ^{14}C. A letter together with a questionnaire was sent out to a contact person at each company (see Appendix B). In total 35 contact persons were contacted at five pharmaceutical companies, twelve biotechnology companies, three universities, three nuclear power plants, six hospitals and six other, and of them seven responded positively. The contact persons handed out the questionnaires to the concerned people who were given a choice if they were willing to donate a few pieces of hair. If the subject agreed, a sample was collected from the back of the subject's head, and sent to Lund University together with the person's questionnaire. In the questionnaire the subject was to answer such things as age, sex, professional category, type of diet such as a vegetarian, type of ^{14}C preparations etc., factors that may affect the outcome of the result.

To see if the persons in this investigation are internally contaminated, the amount of ^{14}C in uncontaminated hair needs to be known. This is later to be subtracted from the value acquired from the hair samples. Small differences may appear due to dietary reasons since the amount of ^{14}C in a humans body are reflected of one's diet.

Hair samples were collected at hairdresser's, to act as a measure of the background in people not involved in the kind of work as mentioned above.

3.2 AMS Measurements

When reporting results in journal or otherwise and calculations, rather than writing the actual amount of ^{12}C, ^{13}C or ^{14}C it is corrected for decay, machine instability, fractionation, contamination etc. and then related to the NBS oxalic acid. A fractionalisation term, $\delta^{13}C$, comes to use because when ^{14}C is taken up by plants, it is affected by isotopic fractionation. Isotopic fractionation means that lighter nuclides like ^{12}C and ^{13}C is preferred to be taken up by the plants because they are lighter. Thus the amount of ^{14}C in the atmosphere will not be exactly the same as the amount of ^{14}C in plants or in animal tissue. $\delta^{13}C$ is measured on all samples as well as on standard samples such as the NBS oxalic acid.

One parameter that is always reported is the *percentage modern carbon* in a sample [20]. This is defined according to (1.1):

$$pMC_{\text{sample}} = \frac{\left(^{14}C/C\right)_{\text{sample}} - \left(^{14}C/C\right)_{\text{fossil}}}{\left(^{14}C/C\right)_{\text{standard}} - \left(^{14}C/C\right)_{\text{fossil}}} \times 100\% \times pMC_{\text{standard}} \qquad (1.1)$$

where:

$\left({}^{14}C/C\right)_{\text{sample}}$ is the measured isotope ration of the sample, corrected for $\delta^{13}C$

$\left({}^{14}C/C\right)_{\text{fossil}}$ is the measured isotope ratio of a fossil sample, corrected for $\delta^{13}C$, to serve as a background of the sample preparation and AMS measurement.

$\left({}^{14}C/C\right)_{\text{standard}}$ is the measured isotope ratio of a standard corrected for $\delta^{13}C$.

$pMC_{standard}$ is the pMC of $\left({}^{14}C/C\right)_{\text{standard}}$ as stated by the supplier.

The supplier of the standard is NBS, National Bureau of Standards and is called Oxalic Acid I. There are standard samples to measure on, when calculating pMC. Two of those standards come from NIST (National Institute of Standards and Technology), and are Oxalic Acid I ($C_2H_2O_4$) and Oxalic Acid II. 95% of the activity of the Oxalic Acid I/II is equal to100 pMC (**p**ercent **M**odern **C**arbon) or to a hypothetically ^{14}C-activtiy from the year 1950.When measuring with SSAMS, a software that takes care of the calculations to pMC. The software is supplied by National Electrostatics Corporation (NEC), Middleton, Wisconsin, USA.

When estimating the excess amount of ^{14}C in an individual from ^{14}C contamination, as stated, the background value must first be subtracted. To be certain that it is a contamination, the calculated value must be at least 2σ over the weighted mean background value. It is important to know that this value does not tell anything about the amount of ^{14}C in any source organ. One might want to continue the research and check contaminations in urine, and make biopsies etc. to make a more accurate estimation of the excess amount of ^{14}C in workers.

3.3 Graphitization

The hair samples were prepared for AMS analysis by graphitization. A suitable amount of hair for the AMS system is about 5-10 mg of hair. When prepared the samples, 5-10 mg of hair was taken out from a larger sample and prepared. When the hair samples were collected it was asked that all samples was collected from the same position on the scalp and cut off with a clean scissor. Especially among the background samples, the hair was analysed several times to see if there was any variations between the samples from the same batch.

The hair samples are washed in ethanol to wash away any external contaminations, and then put in quartz test tubes, to be covered with copper oxide (CuO). The CuO is there to give off more oxygen when the samples are heated, to ensure that the hair sample becomes carbon dioxide instead of carbon monoxide. The tubes are heated in a torch flame to combust the sample, and then gas is passed through an intricate vacuum system, seen in figure 3A and 3B, to get rid of other impurities. In the tube where the carbon is materialised contains a small amount of iron powder that functions as a catalyst. Together with H_2 gas the CO_2 is converted into solid graphite and water. The water is absorbed in an adjacent tube filled with $Mg(ClO_4)_2$. A more detailed description of the process can be seen in Appendix C.

Normally the process of graphitization takes about 4 hours, but when doing the hair samples for background measurements, the process went unusually slow after processing a few of the hair samples. Instead of taking about 4 hours it took about 3 days. The delay may be caused by some other substance released when burning, that would gradually build up on the surfaces of the graphitisation system and obstruct the graphitisation procedure. Another hypothesis

was that new burner was used with a higher flame temperature and would release other types of impurities than previously. Hair is known to contain a significant proportion of sulphur, an element known to inhibit the graphitization process. To take care of this problem, silver wool was put in, absorb any sulphur impurities in the sample. Also a thorough cleaning of the system was made, by taking it apart and cleaning it with ethanol and swabs. New test tubes for the iron powder was purchased, a more careful burning of the sample was made. However, the problem persisted and a new approach was needed.

Therefore a cleaning system was created, that was applied just after the burning unit. This is made by having a quartz tube that have holes in each end, filled with CuO, and stuffed with silver wool at each side. The tube is connected to the system via two couplings and an oven surrounds the quartz tube. Cold traps are placed on each side of the vial to be able to transport the gas through the CuO-trap. The CuO is warmed by the oven in presence of streaming hydrogen gas to remove the oxygen from the CuO. Later when the contaminations come through the system it binds to the Cu. Deionised water is also added to solve the sulphur in water, which can be removed by the cold traps. Tests were performed on different samples of the same hair in order to investigate which of the substances – or which combination of the substances – that were the most effective cleaning agents. The results of the tests can be seen in the table 1 below.

Table 1. Results of graphitization depending on type of chemicals in the combustion tubes. $\Delta p/p(CO_2)$ is the ratio between the pressure difference in the graphitization tubes between t_0 and t_{final} and the initial CO_2 pressure. For a complete reduction of all CO_2 the ratio should be around 3. Unit A, B, C and D denotes each of the four graphitization unites on the graphitization system.

Unit	**Water [≈0.5ml]**	**CuO [≈5.71g]**	**Cu [≈0.5g]**	**Ag [≈0.5g]**	**$\Delta p/p(CO_2)$**	**Time [h]**	**Results**
A	X	X	X	X	3,4	5	GOOD
B	X	X	X	X	2,8	5	GOOD
C		X			0	>48	DO NOT WORK
D		X			2,4	>48	TOO SLOW
A		X		X	0	>20	DO NOT WORK
B	X	X		X	2	>20	SOMEWHAT SLOW. TO SMALL AMOUNT OF H_2 ADDED
C		X	X		1,2	>20	TOO SLOW

D	X	X	X		0	>18	DO NOT WORK
A		X		X	0	>20	DO NOT WORK
B	X	X		X	0,7	>20	TOO SLOW

All of the samples were drawn through the Cu+Ag oven with a temperature of 500°C five times with the help of cool-traps on each side of the oven. To keep everything cooled and only thaw the CO_2 with the help of cool ethanol one could transport the gas back and forth. From these trials one can deduce that one need all of the components to make it work within a short amount of time.

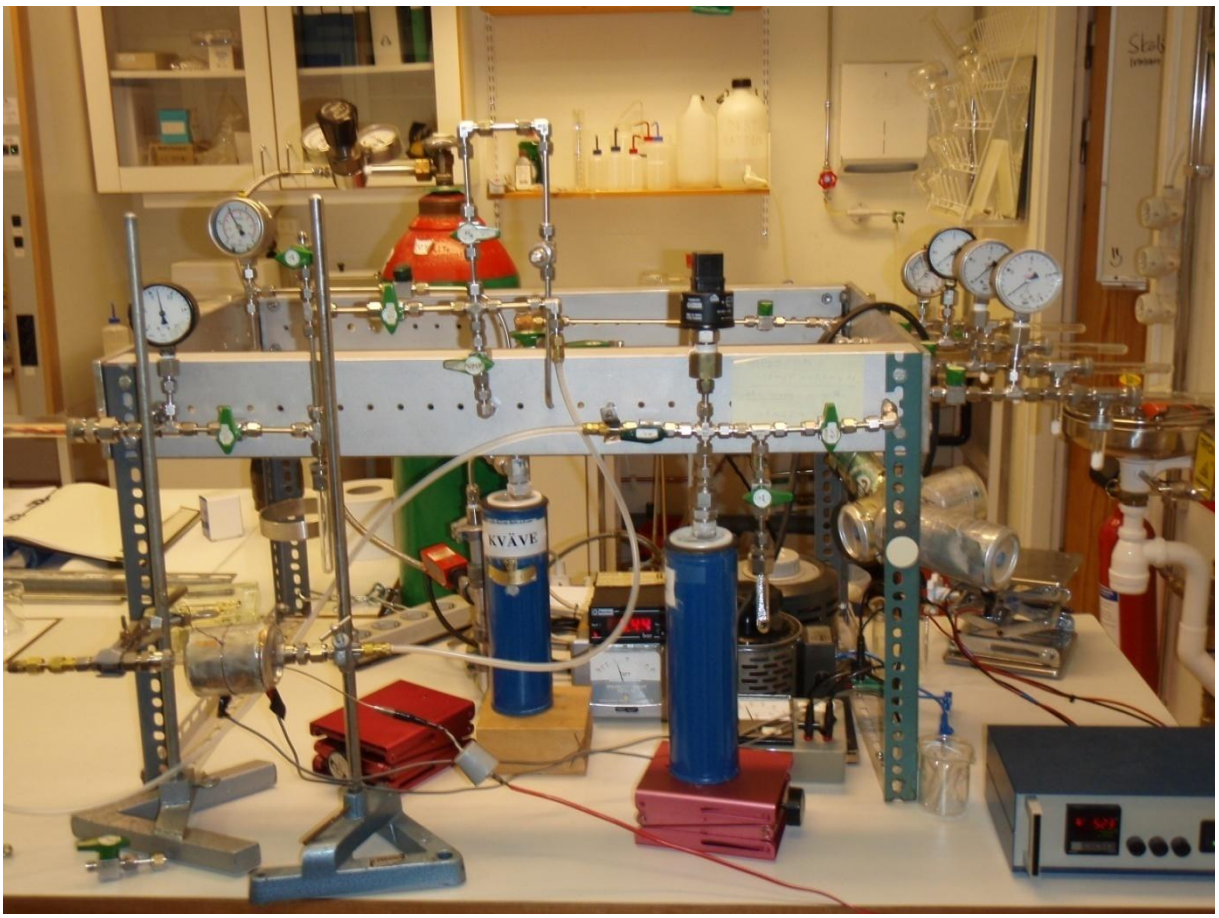

Fig. 3A. The graphitization system, with ovens, pumps etc. The sample is burned with a gasflame at the far left in the picture and then passed through a series of cold traps. The Cu-Ag oven can be seen being held by two stands. At the far right there are tubes where the CO_2 is converted into solid graphite. A sketch of the system can also be seen in figure 3B.

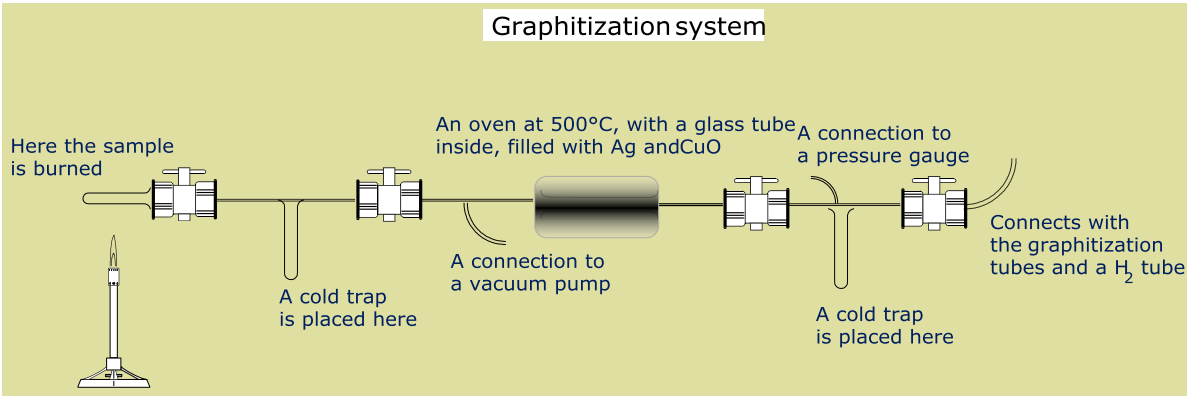

Fig. 3B. A sketch of the graphitization system, where the sample is burned in the left part of the picture and then transported through the system to be cleaned. The CO_2 gas is transported five time through the oven.

From table 1 above, it can be concluded that one have to have all ingredients to make a sample work the best.

3.4 AMS Measurements

The graphite is pressed into sample holders and a small amount of silver powder is mixed in at the end. This is for the heat that is generated in the ion source (when the Cs beam hits the carbon sample) to dissipate more easily. Each sample holder is inserted into the wheel (se figure 4), which is later put into the ion source, and vacuum is pumped. Before the measurements start, a calibration of the beam is essential to get a good result. This involves tuning of the magnets etc. A run list also needs to be set up, where the number of runs, the length of each run, sample name, etc. is put in. When all is done, the measurements start, with a pre-run to see a preliminary result of each sample. If this is good, a more deep analysis of each sample is done, with more time on each sample, and more runs on each sample is done. The results in then logged in a file to be printed out and then analysed using the theory in chapter 3.2.

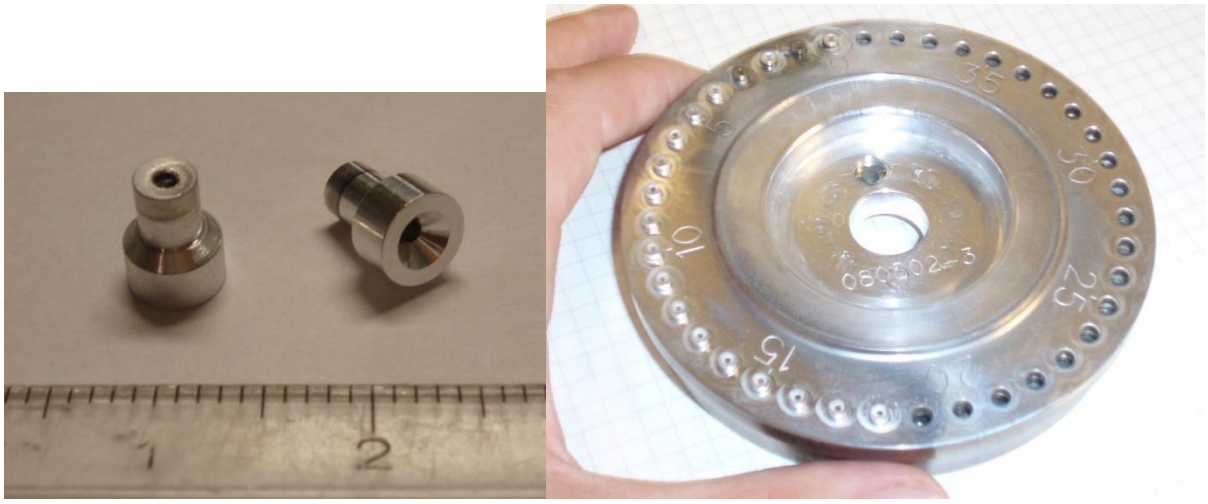

Fig. 4. Here the sample wheel can be seen together with each small sample. The wheel is to be inserted into the accelerator.

4. Results

4.1 Contaminations in hair

The results of the measurement on background samples, i.e. the hair samples collected at hair dresser saloons can be seen in figure 5. There can be seen a small variance from sample to sample. This can be due to environment and/or diet. The weighted average background is 104.8 pMC±1.1pMC. Since there is an natural variance within the background, to be sure that it is a contamination, only values 2σ over the background is considered to be contamination. When comparing coloured hair and uncoloured hair there are no difference in its background value. Coloured hair are samples 12 and 13.

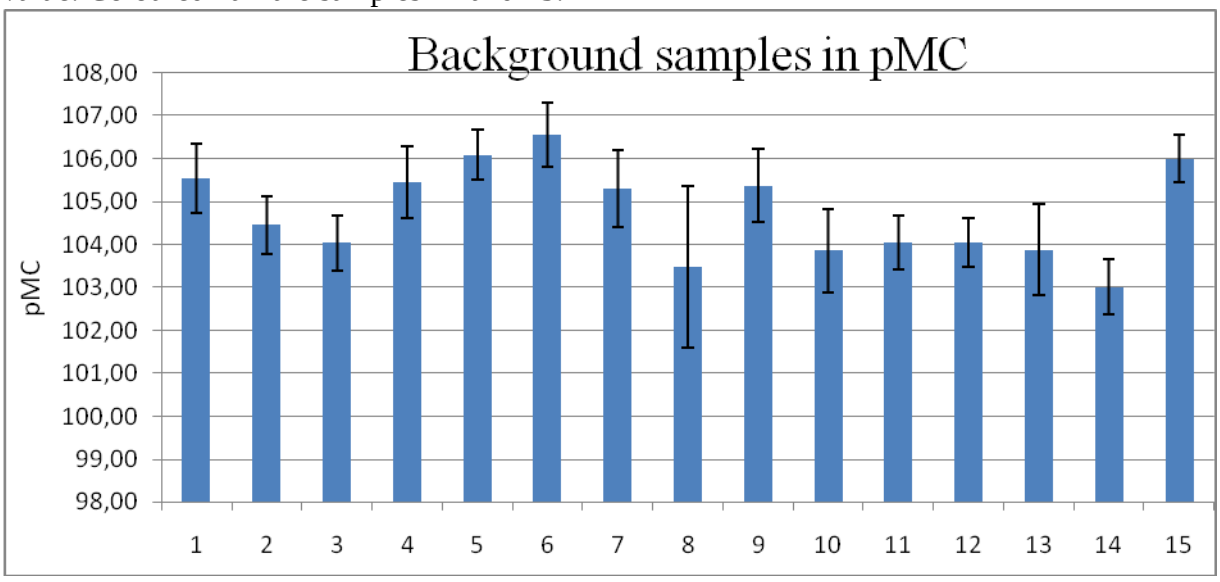

Fig. 5. As can be seen in this figure there is only a little variation of the pMC in the background sample as is to be expected.

The results of the ^{14}C measurements in hair from workers at industry and nuclear power plants is shown in figures 6A, 6B and table 2A and 2B. The result shows contamination in 15 individuals. They are daily users of $^{14}CO_2$, ^{14}CN and other small molecules, but there are also persons involved in the nuclear industry. The highest values are about 10 and 80 times higher than the normal background. One of the workers (the analytical chemist) work in the same place as the two highest contaminated persons, but the person is not involved in ^{14}C-work. The contaminations in workers from the nuclear industry have quite low levels contaminations. In figure 7A one sees all the different persons that were analyzed, and in figure 7B one only sees the one with lower contaminations. Each individual contamination can be seen in table 3A and 3B below.

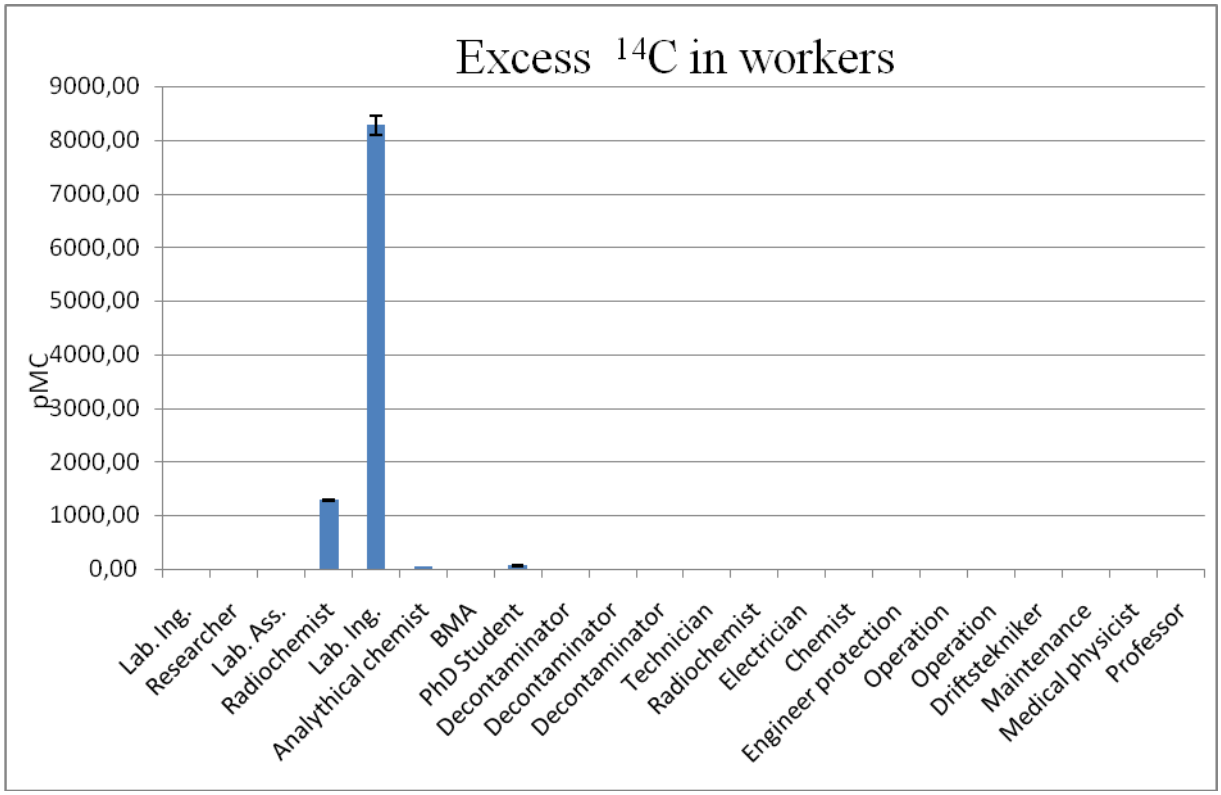

Fig. 6A. The pMC values with background subtracted that each worker have.

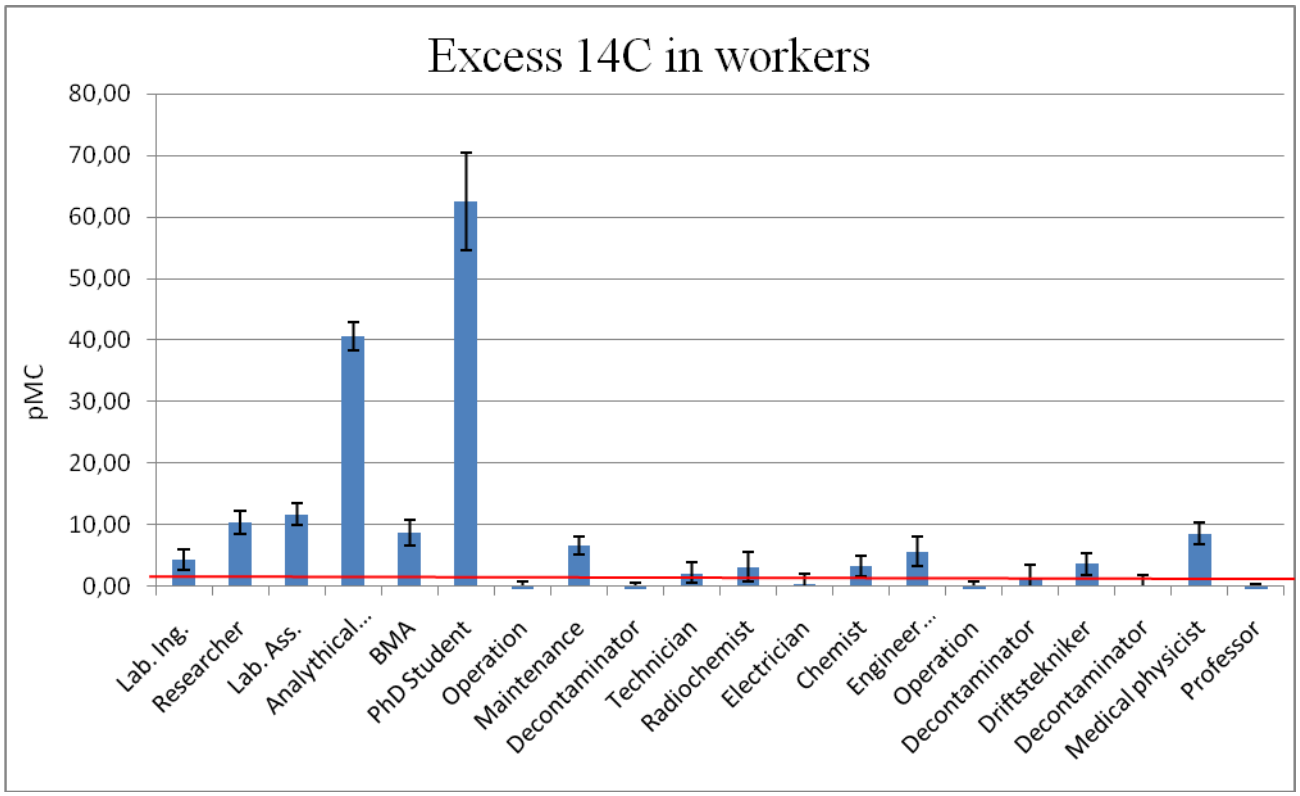

Fig. 6B. Shows the same results as in figure 6A, but without the more extreme samples. Some of these are within the same workplace and that suggests there is a general contamination at the workplace. The red line is the 2σ line, where the samples are considered to be contaminated.

Table 2A. Each person's pMC value and what substance they handle can be seen below. NP: Nuclear power plant personnel, and each number represents a workplace.

Occupation and workplace	**pMC_{korr}**	**Gender**	**Substances**	**Activity [MBq/year]**
Lab. Ing. 1	4.37±1.73	Female	Acrylamid	0.00074
Researcher 1	10.43±1.86	Female	Acrylamid	0.00074
Lab. Ass. 1	11.76±1.80	Female	Mannitol, Acrylamid	18.5
Radio chemist 2	1293.59±13.56	Male	CO_2, CN	74000
Lab. Ing. 2	8281.41±173.35	Female	Small molecules	NA
Analytical chemist 2	40.64±2.28	Male	-	-
BMA 3	8.67±2.10	Female	Protein	0.0370
PhD Student 4	62.56±7.99	Male	Different Sugar types	40.0
NP: Decontaminator 6	1.33±2.17	Male	-	-
NP: Decontaminator 6	0.13±1.61	Female	-	-
NP: Decontaminator 6	-1.11±1.64	Male	-	-
NP: Technician 6	2.17±1.64	Male	-	-
NP: Radiochemist 5	3.21±2.36	Male	-	-
NP: Electrician 5	0.45±1.61	Male	-	-
NP: Chemist 5	3.28±1.62	Male	-	-
NP: Engineer protection 5	5.63±2.37	Male	-	-
NP: Operation 5	-0.74±1.43	Male	-	-
NP: Operation 5	-0.71±1.58	Male	-	-
NP: Driftstechnitian 5	3.70±1.77	Male	-	-
NP: Maintenance 5	6.62±1.48	Male	-	-
NP: Radiation physicist 5	8.53±1.76	Male	-	-
Professor 7	-1.25±1.61	Male	-	-

Table 2B. Describing how much they work in their respective environments, and also if there are any special comments related to each person. Also here each number represents each workplace.

Occupation and workplace	**Time working in 14C environments**	**Comments**
Lab. Ing. 1	2-3 times per year	4 weeks vacation
Researcher 1	2-3 times per year	
Lab. Ass. 1	2-3 times per year	4 weeks vacation
Radio chemist 2	Every day	
Lab. Ing. 2	Once a week	Dyes hair
Analythical chemist 2	-	Works in ^{14}C laboratory but not with the nuclide
BMA 3	Once year	Dyes hair
PhD Student 4	Once week	
NP: Decontaminator 6	Every day	
NP: Decontaminator 6	Waste management every day	Dyes hair
NP: Decontaminator 6	Waste management every day	Special diet
NP: Technician 6	Working with radioactive waste	
NP: Radiochemist 5	40% in controlled area	
NP: Electrician 5	7 weeks a year	
NP: Chemist 5	5-6 months a year, chemistry	
NP: Engineer protection 5	600 h a year	
NP: Operation 5	4-5 months in controlled area	Dyes hair, uses hair products
NP: Operation 5	-	
NP: Driftstechnitian 5	Every day	
NP: Maintenance 5	Mechanical maintenance every day	
NP: Radiation physicist 5	40% of work time	Uses hair products
Professor 7	Subject of an experiment with ^{14}C	

5. Discussion

As seen in the results some of the participants are contaminated with ^{14}C. It may be that some of the excess come from external contamination of the hair, but I think that most of the external contamination is washed away with the ethanol in the preparation step. Also that one of the workers in the same place has contamination in the hair may be an indication that there is a general contamination in the workplace. There are also a small excess of ^{14}C of some other workers that also may indicate a general contamination of ^{14}C. Some possible ways of contaminations can be deduced from the substances and activity they use at each laboratory/workplace, seen in table 2A. Most of the contaminations are probably from inhalation and ingestion, since many of the ^{14}C substances are in gaseous form. In the workplace where a worker was contaminated that was not using ^{14}C, the contaminations must first and foremost come from ingestion.

One can also conclude when graphitization is done with hair, one need all of the cleaning steps in order to make the sample into solid graphite within an acceptable time limit. One can also conclude that all the cleaning steps are needed to take care of the sulphur contamination and that these steps take care of the contaminations very well. One can also see that there is no difference in pMC value from the same hair samples that was made before the graphitization process slowed, compared to after it slowed.

The ones that were contaminated, I would suggest that one would move on and do measurements on their blood and faeces to really quantify how contaminated they are.

Contaminations may arise from a number of different areas, such as the laboratory and its furnishings, the air and the tools or vessels used in a laboratory handling ^{14}C-labelled materials. Contamination is often found on door knobs, water faucets, instrument lids etc. A surface that is often used in work may also be contaminated over the time, because samples or other utilities may carry undetectable contamination. So the contamination builds up over time. Unremembered or unreported spills of ^{14}C, may also leave small contaminations, however thoroughly cleaned. It is also important that personnel from hot labs should not enter the natural labs and vice versa.

When it comes to contaminations in the graphitization laboratory, ^{14}C substances isn't handled in the laboratory. And all samples that are put into the system, that one suspects may contain large amounts of ^{14}C, are first controlled in another separate system, for two reasons. One do not want to have to much signal in the AMS machine, and also to be sure that it's not to much to contaminate the system.

AMS is a good tool use in since it is fast and accurate, but it is also costly, about 750 SEK per sample in Lund. But ^{14}C screening with AMS or any other detector has not been deemed interesting before, because ^{14}C hasn't been thought of as a risk. If that would change cannot be estimated.

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