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AMS and Climate Change

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Abstract

This paper attempts to draw a connection between information that can be gained from measurements with accelerator mass spectrometry (AMS) and the study of climate change on earth. The power of AMS to help in this endeavor is demonstrated by many contributions to these proceedings. Just like in archaeology, we are entering a phase of an 'integrated approach' to understand the various components of climate change. Even though some basic understanding emerged, we are still largely in a situation of a phenomenological description of climate change. Collecting more data is therefore of paramount interest. Based on a recent suggestion of 'geo-engineering' to take out $CO₂$ from the atmosphere, this radical step will also be briefly discussed.

1. Introduction

Climate change is perhaps the most discussed topic in all of science in our time. It has become clear that man's influence in certain sections of our environment is not negligible any more. Therefore Paul Crutzen, the Nobel Laureate in Chemistry of 1995, suggests that the Holocene epoch is over and we have entered the Anthropocene, an epoch of unprecedented influence of man on the earth system. As much as we wish to preserve the natural beauty of our planet, we are in fact taking an ever growing influence on the very habitat we want to preserve. The most obvious effect is the $CO₂$ increase in the atmosphere, shown in Fig. 1. David Keeling started this life-long effort to monitor $CO₂$ in 1958 [1], which eventually became one of the most important results of so-called Cinderella science [2]. It is well established that the atmospheric $CO₂$ concentration increased from 314 ppmv in 1958 to 387 ppmv in 2009. Most likely this is due to burning fossil fuel by man. Much less is understood about the consequences of this increase on climate. Suggestions of how to go about this situation are numerous, and go far beyond scientific discussions. Even though most of these efforts are sincere, they nevertheless often forget how limited our understanding of the climate system actually is.

Fig. 1. Record of the atmospheric $CO₂$ concentration during the past 50 years as measured on Mauna Loa in Hawaii [3]. The measurements were pursued for almost the entire length of time by the late David Keeling from Scripps [2]. Therefore this plot is frequently called the 'Keeling curve'. The seasonal minima are due to the increased uptake of $CO₂$ by vegetation during summer time (growing season).

In order to prepare for the topic of this paper I visited Wally Broecker at the Lamont Doherty Earth Observatory of Columbia University New York. Wally studied the earth system since more than 50 years and likes to picture the climate as a sleeping dragon (Fig. 2). He also describes our understanding of the climate in the following way: "As I sometimes tell my students, the folks in the back room who designed our planet were pretty clever. We have clear evidence that different parts of the earth's climate system are linked in very subtle yet dramatic ways. The climate system has jumped from one mode of operation to another in the past. We are trying to understand how the earth's climate system is engineered, so we can understand what it takes to trigger mode switches. Until we do, we cannot make good predictions about future climate change."

The Sleeping Dragon

Fig. 2. "The complex climate is like a sleeping dragon which should not be disturbed without good reason. But if the dragon wakes, we have to know how to pout him back to sleep again." – Wally Broeker.

So, what can we do about this situation? Perhaps, two points can be made:

- (1) We have to collect much more data from all domains and time periods on earth, in order to test climate models more rigorously and consequently allow more reliable predictions.
- (2) We somehow have to come to grips with the question whether we want to preserve the climate in its current state or whether we want to adapt to the inevitable climate changes – human-caused or otherwise - as life on earth always did in the past.

AMS can contribute a lot to the first point, and I will attempt a personal summary of this. The second point is a sensitive and complicated issue, where roughly three groups with divergent opinions about the human influence on climate can be distinguished: Alarmists, deniers, and pragmatists. In the context of the present paper, only a pragmatist's approach of fixing the climate [4, 5] will be briefly discussed.

2. Radiocarbon in the atmosphere

Radiocarbon dating was developed by Willard Libby in the late 1940s [6-8], and earned him the 1960 Nobel Prize for Chemistry. In the sixty years since its first application for dating [8] three major developments took place. (i) In the late 1950s it was recognized that the ${}^{14}C$ abundance of atmospheric $CO₂$ was not constant in time [9], which required a calibration to obtain reliable absolute dates. Eventually this resulted in highly precise calibration curves mainly based on tree rings reaching back to about 12,500 years [10]. Beyond this time range the uncertainties increase, and great efforts are under way to improve the calibration back to the limits of 14 C dating (~50,000 years). This has been called the 'final frontier' of accurate dating with radiocarbon [11]. (ii) A second important development was the invention of accelerator mass spectrometry in the late 1970s [12-14], which reduced the required carbon sample size by a factor of 1000 and more (grams to milligrams and below). This latter development was also very important for applications connected to climate change (e.g. sections 3), where large sample sizes are either not available at all or very impractical and costly. (iii) Finally, an unintended side effect of the nuclear weapons testing program was the excessive labelling of atmospheric $CO₂$ with man-made ¹⁴C, allowing one to gain unique insight into the dynamics of the global carbon cycle, and related processes.

Fig. 3. Schematic presentation of the processes leading to the production and distribution of $14\tilde{C}$ on earth [15]. The sudden increase of $14\tilde{C}$ in the atmosphere by nuclear weapons testing in the early 1960s is also indicated. The two-step oxidation process is discussed in the text.

2.1. 14CO and the OH radical

An important insight for atmospheric chemistry was the relevance of the OH radical in the oxidation of CO to $CO₂$, and other trace gasses [16, 17]. The exceedingly strong oxidizing power led Paul Crutzen to call the OH radical the "detergent" of the atmosphere. As depicted in Fig. 3, the oxidation of cosmic-ray produced ${}^{14}C$ atoms proceed first to ${}^{14}CO$ via reactions with O_2 , and in a second step to ¹⁴CO₂ via reactions of ¹⁴CO with OH. The instantaneous production of ${}^{14}CO$ from the quasi constant flux of cosmogenic ${}^{14}C$, allowed one to use the ¹⁴CO equilibrium concentrations in the atmosphere to gauge OH concentrations, which are highly variable (see box insert in Fig. 3). A series of AMS measurements of ${}^{14}CO$ air samples from the high-altitude observatory at Mt. Sonnblick in the Austrian Alps revealed seasonal variations of ¹⁴CO coupled to the availability of OH [18]. It was also possible to backtrack the movement of air masses in this way.

Fig.4. The ¹⁴C bomb peak: Deviation of ¹⁴C (Δ^{14} C) from the natural (reference) value in atmospheric $CO₂$ during the second half of the $20th$ century in the northern hemisphere (NH) and in the southern hemisphere (SH) [19]. After the Nuclear Test Ban Treaty (NTBT), 14 C rapidly distributes to other domains on earth (cf. Fig. 3). The slow radioactive decay of ${}^{14}C$ is insignificant for the shape of the bomb peak curve after 1963, which is determined by the mean residence time of ${}^{14}CO_2$ molecules in the atmosphere (~7 yr).

2.2.The 14C bomb peak

Atmospheric nuclear weapons testing in the 1950s and early 1960s increased the natural 14 C content in the atmosphere by approximately an equal amount of anthropogenic ^{14}C . After the Nuclear Test Ban Treaty of 1963 this \sim 100% excess of ¹⁴C got quickly distributed into the other 14C archives on Earth (Fig. 4). In 2009 it has almost reached the pre-nuclear level. Since the ¹⁴C bomb peak has been measured in atmospheric CO_2 since the mid 1950s in both the northern and the southern hemisphere [19], it provides a unique calibration curve reflecting its distribution into the ocean and the biosphere. The importance of bomb ${}^{14}C$ for studying the dynamics of the atmosphere and the oceans is obvious. The latter is being discussed in the contribution of Bob Key to these proceedings [20].

A very different application of the ${}^{14}C$ bomb peak is discussed in the contribution of Kirsty Spalding to these proceedings [21]. Here, the fact that all humans living through the second half of the $20th$ century are labelled with bomb-¹⁴C is being used to determine the age of cells in the human brain and the human body $[22, 23]$. Such ^{14}C measurements are only possible with AMS, but also most of the oceanographic investigations benefited from AMS enormously (see below).

3. Oceanography

3.1. Radiocarbon

Water is the most important ingredient for life as we know it, and the oceans provide plenty of it. Besides being important as the ultimate supply of fresh water and also of all ice on earth, the oceans play an important role in earth's climate through global currents, some of them transporting heat from the warmer, aequatorial regions to the cooler, higher latitudes. Measurements of radiocarbon in ocean water have contributed prominently in studying global ocean currents. During the 1970s radiocarbon was measured in the world oceans within the GEOSECS program (Geochemical Ocean Section Study). This was the pre-AMS period and beta counting of 14 C required the sampling of 250 liters of sea water [24-27]. Among other insights it led to the development of the intriguingly simple model of the Great Ocean Conveyor Belt by Wally Broecker [28, 29], displayed in Fig. 5.

The development of ^{14}C measurements with AMS allowed one to reduce the sea water sample size to 0.5 liters, opening up the possibility of a much more detailed study of the world oceans. The National Ocean Sciences AMS facility was set up in Woods Hole [30], and well over 13,000 water samples were measured within the World Ocean Circulation Experiment (WOCE) project in the 1990s [31]. The enormous data set of both ${}^{14}C$ and $\delta {}^{13}C$ values acquired at Woods Hole is a good example what AMS can contribute to learn more about the dynamics of the world oceans and their interaction with the atmosphere. It's now up to the oceanographers to organize and interpret the data, and then to challenge the modellers to describe the dynamics of the oceans according to these findings. The development in this direction is discussed in the contribution of Bob Key in these proceedings [20].

Fig. 5. The great ocean conveyor logo (Broecker 1987). Illustration by Joe Le Monnier, Natural History Magazine. It is a simplified picture of the main ocean currents transporting heat around the globe. For a more complex picture see Rahmstorf [32].

3.2 Other oceanographic tracers: 39Ar, 129I, 99Tc, 236U, 237Np

Although radiocarbon provides as yet the most detailed information, for a more complete characterization of ocean dynamics [32] other radioisotopes with different properties and sources would be desirable.

A particularly useful cosmogenic radioisotope is 39 Ar, since its half-life (269 y) compares well with the time range of ocean currents $(\sim 100 \text{ to } 2000 \text{ years})$, the atmospheric content is well defined, and its dissolution in surface waters is straightforward. However, atmospheric argon has an exceedingly small isotope ratio $(3^9Ar^{40}Ar = 8x10^{-16})$ and cannot be measured with tandem AMS. Argon forms only metastable negative ions with a mean lifetime of 250 ns [33], too short for acceleration in a tandem accelerator. Although a successful dating of deep ocean water with 39Ar AMS was performed at the ATLAS linear accelerator at Argonne [34], the formidable technical challenges – particularly the suppression of the omnipresent stable isobar $39K$ - prevent its practical use as an oceanographic tracer so far.

Anthropogenic releases of fission fragments into the ocean are being pursued for some time as quasi-stable oceanographic tracers with a reasonably well-known input in terms of spatial and temporal distribution. Distribution of 129 I in Northern Seas have been studied by several groups [35-38]. The abundant fission product 99 Tc is also being considered as a useful oceanographic tracer, and AMS methods for its detection are being developed [39-42]. Another interesting oceanographic tracer would be ²³⁶U, a by-product from the fission of ²³⁵U

since 15% of the thermal-neutron captures on 235 U lead to 236 U. AMS is well suited to measure ²³⁶U/²³⁸U down to natural levels of 10^{-12} [43], Since ²³⁶U/²³⁸U levels in spent nuclear fuel are in the range of 10^{-4} to 10^{-3} , a large dynamic range as a seawater tracer would be available [44]. However fallout from nuclear weapons testing complicates detection at environmental levels [45]. Finally, anthropogenic 237 Np has also been investigated with AMS in seawater [46]. One can expect interesting applications from the above mentioned tracers in the years to come, and AMS will most likely play a prominent role in it.

4. Milankovitch and the Ice Ages

The waxing and waning of the big ice sheets on earth is clearly the most obvious signature for global climate change. Therefore, understanding what drives glacial cycles was always high on the agenda of geoscience in the last century. A big step forward was the development of the astronomical forcing theory of Milutin Milankovitch in the first half of the $19th$ century [47, 48]. In essence it states that the coming and going of ice ages is caused by the change in solar insulation on Earth due to the variation of Earth's orbital parameters. Fig 6 shows the three orbital parameters on which Milankovitch' theory is based.

Fig. 6. Schematic presentation of the orbital parameter of the earth, which vary with the indicated periods. The figure was adopted from the work of Broecker and Denton [28]. On the basis of these variations Milutin Miankovitch developed his theory of the astronomical forcing of the Ice Ages [47].

It took a while before the seminal work of Hays et al. [49] on deep see sediment cores firmly established these variations as the pacemaker of the Ice Ages. The Milankovitch theory is very attractive since it is one of the few climate-relevant processes which is based on solid physical grounds. In addition, it also conveys a similar simplicity as the Great Ocean Conveyor Belt, but also has limitations.

5. Ice Core Research

Ice core research started in the late 1960s, and the first use of $\delta^{18}O$ as a climate (temperature) indicator in ice was the analysis of a ~100,000-year ice core from Camp Century in Northern Greenland [50]. In the ensuing decades ice core research in both Greenland and Antarctica became one of the most prolific fields to study paleoclimate, as it provides the most detailed footprint of atmospheric and climatic conditions in the past. A seminal work in this respect was the analysis of the Vostok ice core in Antarctica [51], which provided a record of atmospheric gases back to 420,000 years (4 glacial cycles). More recently, the time range was extended to 740,000 years by the EPICA ice core covering eight glacial cycles. [52-54]. It is intriguing that $CO₂$ levels during this long time span ranged from \sim 200 ppmv (glacial periods) to ~280 ppmv (interglacial periods), apparently never reaching the current level of 380 ppmv (see Fig. 1).

In Greenland the record in ice cores doesn't reach back in time as far as in Antarctica, but higher resolution due to higher precipitation rate revealed rapid climate (temperature) variations during the last 120,00 years [55], apparently disappearing during the Holocene. It is probably fair to say that one neither understands the true cause of the rapid changes during the last glacial times, nor the relative stability of the Holocene. There are several hypotheses what might have happened, but it is as yet difficult to pin down a particular one. Ice cores from GRIP [56] , GISP [57] and NorthGRIP [58] provide a wealth of detailed information on climate sensitive parameters during the last 120,000 years, and a new polar ice core project, NEEM [59], is just about to get started. One hopes to get an undisturbed high-resolution record of the Eemian, the penultimate interglacial period (123,000-115,000 years ago).

5.1. Ice core research and AMS

Long-lived cosmogenic radionuclides from the atmosphere are incorporated into ice with precipitation and in occluded air bubbles, allowing one to use them as proxies for climaterelevant parameters. For example 10 Be has been used to follow solar variability for 9000 years suggesting a "causal relationship between solar variability and climate change" [60] . An area of intense research is the transition from the last glacial period to the Holocene, roughly the period from 15,000 to 10,000 years ago. Rapid climate changes characterize these transitions, and isotopic signatures of both stable (e.g. δD , $\delta^{13}C$, $\delta^{15}N$, $\delta^{18}O$) and radioactive isotopes (e.g. 10 Be, 14 C, 26 Al, 36 Cl) help to gain a better understanding of what might have caused these rapid changes. A point of particular attention is the Younger Dryas, a sudden cold spell around 12,500 years ago after a brief warm-up "attempt" (Boelling-Alleroed), just before the final warm-up to the Holocene. Detailed comparative studies of the atmospheric 14 C record from marine sediments, tree rings, and corals with the $\delta^{18}O$ temperature record of ice cores [61] revealed a possible fossil sea water pool at intermediate depth in the Pacific Ocean [62]. Here, precise timing of the evidence from different sources requires a reliable ¹⁴C calibration. Great efforts are on the way to extend the tree-ring calibration beyond 12,500 [63-65] to cover this important period of deglaciation before the Holocene.

6. Climate changes and the Holocene

Compared to the last glacial period with large and rapid temperature fluctuations [55], the Holocene is remarkably stable. However, already in the earliest ice core record from Camp Century [50], one can see warmer and colder periods during the Holocene (Fig. 7).By now, this has been refined through a variety of methods including e.g. dendrochronology, pollen records, 14C and surface exposure dating. It seems however, that the most direct indicator of climate change is the waxing and waning of glaciers in high-mountain regions of temperate zones. Notwithstanding complications through regional effects, a wealth of new information is emerging from AMS measurements tracing these movements during the Holocene.

Fig. 7. First δ^{18} O record from an ice core (Camp Century in NW-Greenland) measured by Willi Dansgaard [50], and reproduced from his recent book [66]. Using $\delta^{18}O$ as a proxy for temperature, the last glacial period gives a pronounced minimum. But also the Holocene shows deviations from the mean, with warmer and colder periods. The numbers refer to Marine Isotope Stages [67, 68].

6.1. Tracing Alpine Glacier movements

When glaciers advance during climatic colder and/or wetter conditions they leave traces which can be dated: (i) When they overrun woody vegetation grown during a preceding warmer climate period at these altitudes, wood might be preserved in the ice and released at later stages when the glaciers recede again. 14 C dating combined with dendrochronology can retrace the climatic conditions at those periods [e.g. 69, 70]. (ii) When glacier advance in rocky terrains, rocks are being scraped off the ground, and eventually form moraines. When glaciers retreat, the moraines are left behind as time witnesses of that advance. Moraines can be 14 C dated from buried organic materials (e.g. trees), and/or from surface rocks of the moraines where the build-up of in-situ produced cosmogenic radionuclides (e.g. 10 Be, 26 Al, 36 Cl) provides another means of dating (surface exposure dating). This method, which was pioneered by Nishiizumi et al. [71] is widely used for geomorphological studies since the 1990s [72].

The CRONOS-Earth project (Cosmic-Ray prOduced NUclide Systematics) is a world-wide effort to systematize in-situ studies with cosmogenic radionuclides [73]. Extensive work on surface exposure measurements in connection with the entire last glacial cycle (115,000 to 11,000 yrs ago) in the European Alps were performed by Ivy-Ochs et al. [74] at the Zurich AMS facility, revealing a wealth of new information on glacial movements during this period. It will be interesting to see how these changes can be compared to the ones observed in the Greenland ice cores [55].

The more subtle climate changes during the relatively stable Holocene are of particular interest to us since they connect more closely to our time. Once we understand the causes of the waxing and waning of glaciers during the past several thousand years, we may be able to factor in the human influence during the last 150 years (since the end of the Little Ice Age,1850 AD). Very recently, an impressive set of data on 10 Be surface exposure dating of moraines of the Mueller Glacier in the Mt. Cook region of the New Zealand Alps were published [75]. It is indeed a remarkable achievement of the AMS technology that moraines of the Little Ice Age could be identified, only about 150 years ago. A comparison of the glacier oscillations from New Zealand with the ones from Central Europe [76, 77] and other parts of the northern hemisphere showed a complex pattern, sometimes being out of phase with each other. It clearly indicates how much we still have to learn to understand the climatic signals in different parts of our earth.

6.2. Climate change and archaeology

It is likely that climate changes had some impact on human development and movements in the past. For example, the temporary settlement of the Norsemen (Vikings) in Greenland coincides with the Medieval Warm Period (800-1200 AD). There is some evidence from ${}^{14}C$ and δ^{13} C measurements that the disappearance of the Norseman from Greenland after 1400 AD was connected to the worsening of the climate [78, 79]. The analysis of plant material at the discovery site of the Iceman Ötzi in the European Alps signalled a possible climate change around 5000 years ago, when the Iceman was firmly locked up at high altitude (3210 m.a.s.l.) in a shallow glacier [80]. It is not impossible that a major mid-Holocene climate change may have happened at that time by a combination of different factors including orbital forcing, changes in ocean circulation and variations in solar activity [81]. Recently, spectacular finds of human clothing and equipment at high altitudes (2756 m.a.s.l.) in the Swiss Alps dated with 14 C seem to coincide with several warmer periods between late Neolithic (~5000 years ago) and the Medieval Warm Period [82].

These are just a few of many evidences slowly emerging by combining information on climate change with archaeological evidence. They are examples for a so-called integrative

approach to archaeology. One may hope that there will be more contribution of this kind to help in sorting out the different aspects of climate change during the Holocene.

7. Fixing Climate

7.1. Alarmists, Deniers, Pragmatists

When climate changes are discussed, three different approaches may be distinguished:

- Alarmists argue that man's influence on our environment is causing the current global warming trend, and catastrophic consequences are imminent.
- Deniers believe that the warming trend is largely due to natural causes, and that man's influence is still minor.
- Pragmatists argue that, regardless of causes, we have to do something about the climate now. Besides reducing the output of anthropogenic greenhouse gases (e.g. $CO₂$, $CH₄$), there are suggestions for 'engineering' our environment at man's will.

The last approach will be briefly discussed in this paper.

7.2. Atmospheric CO2

Some facts about global atmospheric $CO₂$ are listed below. All numbers are approximate, following Einstein's advice: "It's more important to be roughly right than precisely wrong:"

This means that we are increasing the atmospheric $CO₂$ content by approximately 0.5% per year, and fuel resources allow us to keep going like this for at least a hundred more years or so. In spite of great political efforts to reduce the man-made contribution – based largely on the reports of the Intergovernmental Panel on Climate Change [83] – a change in this trend is not yet visible. Depending on one's point of view, it may never happen as long as there is fossil fuel available. On the other hand, we may be forced to act if Broecker's dragon (Fig. 2) suddenly wakes up.

7.3. Controlling the atmospheric CO2 content

Adopting Crutzen's statement in the introduction, we have entered the Anthropocene, the period where men's influence on the environment can no longer be neglected. Early warnings about global warming [84] seem to be confirmed by the IPCC reports [83]. Given this fact, various suggestions of how to actively interact with the environment have been discussed. These efforts have been called 'geo-engineering' [85]. For example, Paul Crutzen suggested stratospheric sulphur injections as a means to enhance the albedo of the atmosphere [86]. His suggestion is partly based on the temperature decrease observed after massive $SO₂$ ejection by the Pinatubo volcanic eruption [85, 87].

Assuming that we really want to limit the $CO₂$ content in the atmosphere by technical means, we have to develop ways to extract it out of the atmosphere and to sequester it permanently. Even though this sounds utopian, it's not impossible [5, 88]. A recent description of these efforts can be found in the book of Kunzig and Broecker [4]. In essence it is proposed to construct carbon scrubber units which take out a ton of $CO₂$ per day from the atmosphere, recover the $CO₂$ from the scrubber, and then sequester it in a safe and permanent way, ideally as $CaCO₃$ or MgCO₃. A simple calculation shows that 400 million units would be needed to take out the 4 GtC per year. This is a big number, but is probably smaller than the number of cars operated on our planet. Technologically it looks feasible, but do we really want to do this?

8. Conclusion

AMS has matured into a technology which measures ultra-low traces of long-lived radioisotope with unprecedented sensitivity in almost every field of our environment at large [89]. Its use goes far beyond climate research, but it clearly became an indispensable tool for studying climate change in the past and presence. An interesting challenge for the future is the use of cosmogenic radioisotope pairs to measure very old ice on Earth. Recent efforts to establish a baseline for this method using the 26 Al/¹⁰Be isotope ratio as a clock in the millionyear age range look promising [90]. The basic idea is that the ratio is less prone to temporal variations of the production of the individual radioisotopes by cosmic rays [91], and thus can be used as a more accurate clock than a single radioisotope. Another radioisotope pair, 36 Cl/ 10 Be, looks also promising for somewhat younger time periods (200-800 kyrs). This dating method has been recently used by Willerslev et al. [92] for deep ice cores in Greenland. This work investigated ancient biomolecules from basal ice, revealing a forested southern Greenland within the past million years.

Another archive which is being used to trace paleoclimate are speleothemes. This is an emerging field, which can give complementary information to glaciers [93]. As there are many caves around the world it is bound to become a rich supplier of information on climate change as well.

At this time, it seems impossible to describe climate change from first principles, since we simply do not understand enough about the climate system as a whole. AMS measurements will certainly continue to contribute in the future prominently to increase our data set. One can hope that eventually a more consistent picture will emerge. Meanwhile we keep watching Broecker's climate dragon, and do what Robert Frost described so well: "We dance 'round in a ring and suppose, but the secret sits in the middle and knows."

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