Sender: M. Allan,
Département de Géologie, Université de Liège
Allée du 6 Août, B-4000 Liège, Belgique
e-mail:mohammedallan@outlook.com

<u>To</u>: Dominik Fleitmann
Editor for Climate of the Past

Object : revised version of paper No: cp-2017-91

September 08, 2017

Dear Professor Dominik Fleitmann

We have the pleasure to prepare for Climate of the Past a revised manuscript entitled " Does Belgian Holocene speleothem record solar forcing? ″ (No: cp-2017-91). We take the opportunity to submit a revised version of our work, including all reviewer comments. All comments (changing, modifying, spelling) suggested by the reviewers have been taken into account in the manuscript. Here are the answers to the comments of the reviewers on the ms. Note the comments are reported in italic and the reply in blue color. We join after the review the modified ms with changes underlined in yellow color.

Hoping that the revised version fits the requirement for publication in Climate of the Past.

Your sincerely

For the authors,
M. Allan

D. Fleitmann (Editor)

The current title "Does Belgian Holocene speleothem records solar foring and cold events?" contains a grammatical error. Unfortunately, we are not able to correct the title at this stage of the review process. We therefore ask the authors to correct the title of the revised manuscript.

The title was corrected "Does Belgian Holocene speleothem record solar forcing?"

Anonymous Referee #1

- *General comment:*

This paper addresses an interesting question, namely whether Holocene climate variability was influenced by past changes in solar activity and if these fluctuations are recorded in a speleothem from Belgium. In general, the paper is well written, the data are interesting and may have the potential to tackle this question. However, in its current form, I cannot recommend the paper for publication in CP. My main concern is that the discussion and interpretation of the presented trace element records is mainly based on correlation and time series analysis. The similarity of the individual trace element signals and their similarity with the d18O record is – in my opinion – not sufficient for a robust interpretation in terms of climate variability. The same is the case for the interpretation in terms of solar forcing. The C1simple observation of cycles with a similar period as solar variability may indicate a solar origin, but to in order to prove this relationship, the authors should at least present a hypothetical mechanism, how the trace element records could be influenced by solar variability. In other words, I would like to see a much more detailed discussion of the potential processes controlling the trace element and stable isotope signals of the speleothems.

We have taken into account this point. The text has been modified. The relationship between trace elements and solar variability is better explained in the new version of the manuscript. See the following section included in the discussion. Here we present a hypothetical mechanism, how the trace element records could be influenced by solar variability. Climate models and paleoclimate proxies (Shindell et al., 2001; Kodera, 2002; Rimbu et al., 2004) have implicated North Atlantic Oscillation (NAO) variability as an important element in the climate response to variable solar forcing. The NAO is known to influence precipitation $\delta^{18}O$ ($\delta^{18}O_p$) through its control on air temperature and on the trajectory of the westerly winds that carry moisture onto Europe during boreal winters (Smith et al., 2016). During the periods of low solar activity, a NAO negative conditions caused a southward displacement of moisture delivery to Europe, and are associated with increased humidity in Central and Southern Europe (Smith et al., 2016; Magny, 2004). This does not necessarily mean that solar forcing was the main source of all Holocene climate variability since different dynamical processes, such as explosive volcanic eruptions, fluctuations of the ocean thermohaline circulation or internal feedbacks, might also have played an important role (Wanner et al., 2008).

Detailed comments:

- *Section 2 provides a nice summary of the potential processes influencing speleothem trace element signals. This shows that the authors are aware of all these complicated processes. I strongly suggest that they discuss their records in terms of these processes, rather than just relying on correlations and frequency analysis.*

The text of the discussion has been expanded. See new paragraph 5.1. $\delta^{18}O$, $\delta^{13}C$ and trace element.

- *Line 114 ff.: The stalagmite was dated in three different labs. In terms of comparability, it would be good to provide a bit more information on the methods used in the individual labs (spike calibration, decay constants, etc.).*

The methods are detailed in the ms as requested.

- *Table 1: Please use points instead of commas for decimal separation. In addition, please provide uncertainties for U content and activity ratios. Have the ages been corrected for this? Is this reflected in the age uncertainties? Please provide more information.*

Both the method section and the table have been corrected.

- *Line 124ff.: Please provide more information on the construction of the age model. Which model was used?*

We use StalAge algorithm, the information and reference have been added in the text.

- *Line 150 and throughout the MS: "Trace element results are closely correlated (r = 0.48-0.77 ..." Please do not provide ranges for r-values, but report the individual correlation coefficients for all records. This information could also be listed in a Table. In addition, how were r-values calculated in case of different temporal resolution of the records?*

The correlation coefficients are now included in the text. To calculate the coefficient r-values we compare the stable isotopes data measured with a sampling resolution of 5 mm with the low resolution dataset for trace elements (also at 5 mm resolution).

- *Line 158: Fig. 5 is mentioned prior to Fig. 4 in the text.*
We check this: Fig.4 is mentioned in line 149 before Fig.5 (line 158).

- *Line 163ff.: "The d18O isotopic composition of the calcite in the Père Noël cave is largely controlled by water availability (drip rate) in the cave. Changes in isotopic equilibrium conditions are driven by the changes in cave humidity and linked to changes in precipitation and evapo-transpiration (Verheyden et al., 2008)." Please provide more information on this. Of course, you do not have to repeat all the details if you refer to published work, but the interpretation of the d18O values is crucial for the interpretation of the trace elements. Thus, at least a short discussion of the major drivers of the d18O signal should be given here.*
The text has been modified with more explanation. See new paragraph 5.1. δ^{18}O, δ^{13}C and trace element.

- *Line 175: "(r(Sr, Ba)=0.77, r(Mg, Sr)=0.72, r(Mg, Ba)=0.48)" This is much better (see above) and should be used throughout the paper.* Ok

- *Line 180ff.: "The positive correlations of Al with Mg, Ba and Sr (r =0.45) suggest that Al content is controlled by the same process than the other trace elements." This is the major problem of the whole paper in its current form. I agree that a correlation may suggest a similar process influencing all trace element records. However, whereas Mg, Sr and Ba are mainly transported as solutes in cave waters, Al is normally transported attached to particles and colloids and thus a proxy for detrital content. Therefore, the correlation of Mg, Sr and Ba with Al – in my opinion – suggests that Mg, Sr and Ba are not only incorporated into the speleothem in dissolved form, but also associated with detrital material. Actually, the positive correlation suggests that the signals are dominated by the content of detrital material. As a consequence, the interpretation of Mg, Sr and Ba in terms of past hydrological variability is complex. For instance, a similarity of these signals has often been interpreted as a result of PCP in the aquifer above the cave. High values would then reflect drier conditions. This could also be proposed for the PN stalagmite. However, high Al content reflecting a high content of detrital material (particles/colloids) would suggest higher flow rates in the aquifer and thus wetter conditions, which would be contradictory. In summary, as outlined in my general comment, the authors must provide a much more detailed discussion of the processes affecting the trace element and stable isotope records. In the current form, all is based on correlations, which can be – as described above – misleading. In this context, it would also be good to see the d13C record if available. As trace elements are strongly influenced by processes in the soil and karst, the comparison with d13C values may provide important additional information.*
We agree with this comment. The text has been corrected. See new paragraph 5.1. δ^{18}O, δ^{13}C and trace element.

- *Line 187ff.: "First spectral analysis demonstrates that trace element time-series contain significant periodicities ..." Maybe I missed that in the paper, but how was the significance of the periodicities determined?* See supplementary data.

- *Line 191 ff.: "Second trace element PN time-series are compared with available temperature record from Mekelermeer core in the Netherlands (Bohncke J., 1991) in order to evidence a regional temperature influence on the PN record." I do not understand why the records are compared with a temperature record if all proxies are interpreted in terms of precipitation. This is a general problem of the paper, in particular in section 5.3. If the records are believed to reflect past precipitation, they should (at least mainly) be discussed in this context. In section 5.3, however, several cold events, such as the 8.2ka event, are discussed as well. Please be more precise here. In addition, it would be better to compare with another record than a relatively old non peer-reviewed record from a PhD thesis.*
Ok. The text has been modified: in particular we remove the comparison with T° record and the reference has been removed.

- *Line 213ff.: "Since the changes of trace elements in the PN speleothem were formerly demonstrated as due to changes in recharge, i.e. effective precipitation, the study suggests that variations in solar activity may be a significant forcing on either the precipitation or on the soil activity or vegetational activity intensity." If trace elements reflect soil and/or vegetational activity, this could be expressed by a similarity with the d13C signal (see above). I really suggest to show and discuss d13C as well.*
Ok, the 13C data have been added in the new version.

- *Line 221ff.: "The higher positive correlations between trace elements concentrations C4(r=0.5-0.9) observed during cold periods ..." See above. Please provide more information here. Which signals were correlated? Which periods were used? Why are cold periods studied if the trace elements reflect precipitation?*
The text has been modified. The correlation coefficients between the trace elements Sr, Mg and Ba have been calculated and reported in the text for several intervals that are considered as cold events in the literature. In our data set we found higher concentrations in TM during those intervals. We interpret the observed higher concentrations by drier conditions.

- *Line 234ff.: "The oldest part of the PN stalagmite, from 12.7 to about 12.1 ka BP, is characterized by the lowest contents of trace elements and low d18O, suggesting wet conditions (Fig.4)." Except for d18O (and maybe Ba), I do not see this. Mg and Sr are on a similar level as in many other phases of the Holocene.*
We agree. The sentence has been deleted.

- *Line 260ff.: "The covariance between geochemical proxies support, as explained in Verheyden et al. (2008), their interpretation in terms of dry-wet changes with higher trace elements and higher δ18O values linked to drier conditions." I agree that d18O and the trace element signals show some similarity on the orbital to millennial scale. However, on shorter time scales, and in particular during some of the trace element peaks, d18O values show an opposite behavior (low d18O/high trace elements, Fig. 4). This suggests that the relationship is not as obvious as proposed by the authors. Please provide a more detailed discussion.*
We have to be cautious in the comparison between TE and stable isotopes, especially at shorter time scales due to the different sampling resolutions. Stable isotopes have been measured with lower resolution (5 mm) than TE (0.3-1 mm).

- *Line 324ff.: "Based on trace element time-series we demonstrate that several events at 10.3, 9.3-9.5, around 8.2, 6.4-6.2, 4.7-4.5, and around 2.7 ka BP alternate with periods of relatively stable and wet/warmer climate." Here and throughout the paper: How do you infer warm/cold, if the proxies reflect precipitation? This needs to be clarified, based on a detailed discussion of the processes influencing the proxy signals.*
The text has been corrected. We compare the intervals that we interpreted as drier or wetter with events that are reported in the literature and that correspond to warm or cold climate.

L. Comas-Bru (Referee 2)

This manuscript presents a new set of trace element data from a speleothem from Pere Noel's cave. The study is focused in addressing whether solar variability during the Holocene has been recorded in this speleothem. Even though there is a potential for a very interesting study on trace element variability in speleothems during the Holocene, I cannot recommend this manuscript for publication in its current form.

General comments
My main concern is that the authors do not acknowledge the fact that correlation does not imply causation. The authors base their interpretations solely on correlations and wavelet analysis, without attempting to invoke the physical mechanisms controlling the observed trace element variability and its potential link with solar variability. Therefore, the scientific methods and assumptions used in this study do not support its main claim: "Our study (. . .) emphasizes that speleothem trace element profiles may be considered as a new solar activity proxy on decadal to centennial timescales over the Holocene" (L331-333).
The physical mechanisms controlling the observed trace element variability in the Pere Noel cave were previously studied. See the references cited in the text (e.g., Verheyden el al., 2000, 2001, 2008). This is now included in a new paragraph (see lines 180-225).

Specific comments
-The overall presentation of the paper is structured and clear. In this regard, I would only suggest to merge Figure 4 and 7, as most time-series are shown in both figures.
We prefer to keep the two figures: seven profiles are already presented in Figure 4.

- The English is correct throughout the manuscript (I've listed some typos at the end of this review).
Ok, minor errors have been corrected.

- The significance level of the correlations is not provided for any correlation mentioned throughout the manuscript and therefore the reader does not know whether these are significant or not.

Ok, more detailed are now given in the method section, the correlation coefficient is significant when r values are close to the value 1: $0.5 < r < 1$: strong correlation, $0 < r < 0.5$: low correlation.

- The authors do not use the age model uncertainties in any of their calculations. They are, however, assessing specific periodicities that may not be significant at all if a Monte Carlo approach is used to take include one of the few uncertainties in the speleothem records that we can quantify!

In our study we use the mean age produced by *StalAge*, a program that allows to calculate the range of age at any depth. Figure 2 presents the min, mean and max age for any depth interpolated from StalAge and based on punctual U-Th dates. The age uncertainty, inherent to any paleoclimate record, is influenced by the uncertainty on U-Th dates. Such uncertainty is lower than 71 years (see Table 2) over all the Holocene. Indeed 92% of the time gaps in the original time series are lower than 25 years and the remaining 8% all occur at once at the end of the signal (after 9500 BP). See also supplementary figures 7& 8.

- No information is provided on how the age model has been constructed. This would be useful to understand the reason behind the asymmetric errors for some sections of the speleothem shown in Fig 2.

Ok, more detailed are now given in the method section. StalAge (Scholz and Hoffmann, 2011) was used to interpolate the ages between the U/Th-age points.

- Most of the interpretations are based on wavelet analyses, but information on how they've been constructed is only available in the supplementary data. This should be part of the main "Methods" section.

We prefer to keep wavelet analyses section the in Supplementary data.

- It is not clear how have the authors resampled the trace element data at constant time interval to perform the frequency analyses (has it been with a simple mathematical two point interpolation?). One needs to be very careful with how the interpolation is done as the reconstructed periodogram (spectral power versus frequency for the resampled series) may differ from the original one. And this would make the reconstructed series unsuitable for wavelet analysis (ie frequencies shown in the wavelet spectrum may be just an artefact, predominantly at high frequencies).

Prior to perform the spectral analysis each dataset was resampled as follows : for each age t (in BP) in the evenly spaced sequence 1789, 1799, 1809,..., the corresponding concentration of trace element is obtained through a linear interpolation between two consecutive data points whose ages (in BP) surround t. Since 92% of the time gaps in the original time series are lower than 25 years and the remaining 8% all occur at once at the end of the signal (after 9500 BP), the effects of such a resampling on the results presented are negligible, especially for periods such as the de Vries cycle or larger. See more information in supplementary materials Fig. 6-8.

- Regarding the wavelet analyses shown in Fig.6, the authors mention frequencies that I do not manage to see in the plots. For example, in L189, "the temporal stability of those periodicities is confirmed by wavelet results". I cannot see any predominant band (redish colour) in the top three panels (the trace elements) lasting more than about a century over a millennial length record. Therefore, stability of those periodicities is not proven.

Ok, the text has been modified.

- Along the same lines as the comment above, the 1000-yr band mentioned in L269 is only present in Ba but not in Mg or Sr. At the moment the authors mention this band as if it was present in all panels but this difference deserves some discussion.

Indeed continuous cyclic period of around 1000 yrs are presented during the interval 10.7-10.3 ka BP but with different spectral powers for the different elements. The 1000-yr band is especially pronounced for Ba and Sr than Mg. The text has been modified.

- Also regarding the wavelet analyses, the authors need to be careful not to interpret frequency signals that are outside of the cone of influence, as these may be under/overestimated. See for example, the discussion on the Early Holocene (section 5.3.1, c. L258) where bands outside of the cone are used in the discussion.

No wavelet analyses were discussed in L258. PN speleothem suggests a wet Early Holocene with dryer conditions from 10.7 to 10.3, at 10.0, at 9.7, at 9.2 and from 8.5 to 8.2 ka BP (Verheyden et al., 2014). Trace element contents in the PN stalagmite display significant variability during the Early Holocene, with three maxima observed around 10.5, from 9.5 to 9.2 and around 8.2 ka BP (Fig. 4, 7).

- Considering that the three comments below are the same, why do they mention different correlation coefficients? Also, please consider not repeating sentences this similar. "Statistically significant positive correlations (r(Sr, Ba)=0.77, r(Mg, Sr)=0.72, r(Mg, Ba)=0.48) are found between Sr, Ba and Mg over the Holocene period suggesting either a common or strongly related controlling process." (L175); "The higher positive correlations between trace elements concentrations (r=0.5-0.9) observed during cold periods which also suggest common control factors." (L221); "The positive correlations (r=0.47-0.98) were observed between trace elements concentrations during those periods which confirms common control factors." (in L259).

The correlation coefficients between the trace elements Sr, Mg and Ba have been calculated and reported in the text for several intervals :
-all Holocene period (L175),
- for dry/cold periods that centered at 10.5, 9.3, 8.2, 6.3, 5.4, 4.6 and around 2.7 ka BP (L221)
- for Younger Dryas and Early Holocene(L259).
The repetition has been removed from the text.

- Assuming that the three main trace elements are controlled by similar mechanisms (which most probably do), the following statements cannot be used to interpret dry conditions: "positive correlation (r=0.45-0.95) was observed between element trace concentrations confirming dryer conditions" (in L290); "The highest positive correlation (r=0.78-0.97) observed between trace element concentrations during this period confirms dry conditions" (in L301).

We agree. The text has been modified. A paragraph was added to better explain the processes that controls trace elements in PN cave.

- The authors say that this is a "high-resolution" record, but no information is provided of the temporal resolution between samples. Moreover, "high-resolution" is a vague term that strongly depends on everyone's expertise and background so, I'd suggest the authors to refrain from using it unless extra information is given.

Ok, it is now added in the method section. The age model reveals a growth rate varied between 0.02 and 0.65 mm.yr^{-1}. Consequently, the analysis of a diameter of 50 micrometers each 300-1000 micrometers interval corresponds to a sample of 1 to 3 years every 0.5 to 50 years.

- How would you explain the peak observed at c. 5750 ka BP (figure 4 and 7), which is higher than some of the other peaks interpreted on the basis of Bond's cycles? (see L221).

Indeed the peak is observed at 5500 ka and not at 5750 ka BP. See more details in the text for this period. Between 5.5 and 5.4 ka BP, the trace elements display an anticorrelation with sunspot number suggesting relatively dry conditions (Fig.4). During this period, trace elements present frequencies corresponding to different solar cycles, i.e. the Gleissberg cycle (70-100 yr), Hale cycle (130 yr), de Vries cycle (200-210 yr), 500 unnamed cycle and Eddy cycles (1000 yr) (Fig.6).

- "The larger diameter of the stalagmite compared to its mean diameter suggests that there is enough water to flow down on the flank of the stalagmite and to precipitate calcite" (L236), when? In the lower part at ~12.0 ka. Is this observation enough to interpret a wetter period? Could that precipitation take places many years later?

The stalagmite diameter is rather large compared to its mean diameter. Dreybrodt (1988, 1999) demonstrated that for constant growth rate of a stalagmite, the stalagmite diameter shows a positive relationship with drip rate and thus water availability (see also chapter 7.2. in Fairchild and Baker (2012) for more explanation.

In the discussion about the Younger Dryas, the authors present "humid" and "warm" is if they were the same concept (which they're absolutely not!): "All proxies measured in the PN stalagmite suggest that a humid period occurred between 12.7 and 12.1 ka BP. This is in agreement with the warm climate during the early YD (12.9-12.15 ka BP). . . found in lake sediments from. . ." (from L239 onwards).

We agree. Our chronological and geochemical pattern of the Père Noël stalagmite suggests a humid period. This period is synchronous with warm to mild summer temperatures and the presence of a local vegetation cover during the YD as found in a north-eastern German varved paleolake (Neugebauer et al., 2012).

- In would be helpful for the reader if the caption of Figure 6 explained the meaning of the y-axis as well as the cone of influence. The authors could consider adding an extra panel of Period vs Power for all series (Mg, Sr, Ba, temperature and Sunspot number). This would help comparing the periodicities of the records.

Ok. The caption has been modified.

- L190 and L194: Wavelet analysis is mentioned here as being part of first and third points.
Ok, it is corrected.

Grammatical errors / typos
L89: Treble instead of treble - Corrected
L40: "...and/or...., and..." in a list of items. Please use and just between the last two. Modified
L126: calculated instead of calculated. Modified
L144: ka BP instead of BP ka. Corrected
L205: there are two commas together after et al. Corrected
L301: high instead of highest. Corrected

Interactive comment (J. Vinós)

The manuscript by Allan et al. under review constitutes a very valuable contribution to an important discussion regarding climate evolution during the Holocene, that could greatly benefit from some improvements. It is exclusively with that aim that I write this commentary. The paleoclimatology field is undergoing an interesting controversy over the existence of Holocene climatic oscillations or quasi-cycles for which no clear mechanism has been identified. For some authors this type of periodicities are likely to emerge from the noise generated by a chaotic climate system (Turner et al., 2016). To other authors the climatic periodicities are real even if the forcings responsible are unclear at present (Khider et al., 2014). This manuscript is an important contribution towards cementing the correlation between some of these climate periodicities and solar variability periodicities as recorded by cosmogenic isotopes records. The hydrological nature of the evidence makes it particularly valuable. The good resolution of the speleothem (∼ 20 years on average), and the high correlation between the different trace elements analyzed make this proxy a very good data source to reconstruct the centennial and millennial climatic changes underlying the differential deposition of the elements. The article would benefit greatly from a discussion of known hydrological changes during the period analyzed with a special emphasis on how they could affect Père Noël cave hydrology and trace element deposition. Modeling (Shindell et al., 2001), climate data (Kodera, 2002), and proxy reconstructions (Rimbu et al., 2004) have implicated North Atlantic Oscillation variability as an important element in the climate response to variable solar forcing. Modes of NAO variability are responsible for wind strength and moisture delivery to Europe (Smith et al., 2016). During the periods of lower than average solar forcing discussed in the manuscript, dominant NAO negative conditions caused a southward displacement of moisture delivery to Europe, and are associated with increased humidity in Central and Southern Europe (Smith et al., 2016; Magny, 2004), and decreased humidity in non-coastal Northern Europe (Deininger et al., 2016; Harrison et al., 2002). The hydrology of the Belgian region, as inferred from the Père Noël cave speleothem record, should be compared to the hydrological groups defined by Harrison et al., 2002, based on lake levels, where it probably belongs to group 5. Even more relevant is the comparison to European speleothem δ18O records presented by Deininger et al., 2016, as it is the same type of proxy based on a different chemical species. Bunker cave, located in Germany (51◦N, 8◦W; Fohlmeister et al. 2012) is close enough to merit a comparison that could allow to draw some conclusions on the similitude of hydrological conditions. It is also my opinion that the title of the manuscript has been poorly chosen. Speleothems record essentially hydrological changes, and the data presented does not allow any inference towards temperature changes. The paper should concentrate mainly on hydrological changes during the Holocene. There are plenty of papers on Holocene temperatures while papers on hydrology are not so abundant. I hope my modest contribution is useful both to the authors and the editor of this manuscript.

Thanks for the positive comments. We therefore modify the title by focusing on solar forcing record in speleothems. The text has been modified and includes now more information about the hydrological conditions. In particular we now present the main results from previous cave monitoring (see section 5.3). The drier intervals that we detect in our PN dataset are compared with European climate event reported in literature. In particular we compare TN profiles with the frequency of warm/cold and wet/dry events as defined by Wanner et al. (2014) in the Northern Hemisphere (see figure 7).

Does Belgian Holocene speleothem record solar forcing?

Mohammed Allan[1], Adrien Deliège[2], Sophie Verheyden[3], Samuel Nicolay[2], Yves Quinif[4], Nathalie Fagel[1]

[1] AGEs, Département de Géologie, Université de Liège, Allée du 6 Août, B18 B-4000, Liège, Belgium

[2] Institut de Mathématique, Université de Liège, Allée de la Découverte 12, B37 B-4000 Liège, Belgium

[3] Politique scientifique fédérale, 231 Avenue Louise, Bruxelles, 1000- Brussels, Belgium,

[4] Université de Mons, Rue de Houdain 9, B-7000 Mons, Belgium

Correspondence to: M. Allan (mohammed.allan@ ulg.ac.be)

Abstract. We present a decadal-centennial scale Holocene climate record based on trace elements contents from a 65 cm stalagmite ("Père Noël") from Belgian Père Noël cave. Père Noël (PN) stalagmite covers the last 12.7 ka according to U/Th dating. High spatial resolution measurements of trace elements (Sr, Ba, Mg and Al) were done by Laser-Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS). Trace elements profiles were interpreted as environmental and climate changes in the Han-sur-Lesse region. Power spectrum estimators and continuous wavelet transform were applied on trace elements time series to detect any statistically significant periodicities in the PN stalagmite. Spectral analyses reveal decadal to millennial periodicities (i.e., 68-75, 133-136, 198-209, 291-358, 404-602,912-1029 and 2365-2670 yr) in the speleothem record. Results were compared to reconstructed sunspot number data to determine whether solar signal is presents in PN speleothem. The occurrence of significant solar periodicities (i.e., cycles of Gleissberg, de Vries, unnamed 500 years, Eddy and Hallstat) supports for an impact of solar forcing on PN speleothem trace elements contents. Moreover, several intervals of significant rapid climate change were detected during the Holocene at 10.3, 9.3-9.5, around 8.2, 6.4-6.2, 4.7-4.5, and around 2.7 ka BP. Those intervals are similar to the cold events evidenced in different natural paleoclimate archives, suggesting common climate forcing mechanisms related to changes in solar irradiance.

Keywords: Holocene, climate change, solar forcing, cold events, speleothems, wavelet analysis, trace elements, paleoclimate time-series.

1. Introduction

The Holocene period appears as a relatively stable climatic period compared to Quaternary glacial/interglacial variations. However several high-resolution studies have emphasized many climatic oscillations over the last 11500 yr (e.g. Mayewski et al., 2004; Wanner et al., 2008, 2011). The Holocene climate variability was tentatively attributed to orbital forcing, volcanic, and/or solar activity (Wanner et al., 2008). Many climatic proxies (e.g., $\delta^{18}O$, $\delta^{13}C$, $\Delta^{14}C$, ^{10}Be, REE content, pollen, dust, humification, element traces) have been measured in different geological archives: in peat (e.g., Allan et al., 2013; Marx et al., 2009, 2011; Shotyk et al., 2001, 2002), speleothems (e.g., Fairchild and Baker, 2012; McDermott et al., 2011; Genty et al., 2003; Verheyden et al., 2008), ice cores (e.g., Delmonte et al., 2002; Gabrielli et al., 2010; Thompson et al., 2000), and in lake sediments (e.g., Magny M., 2004), to reconstruct the past climatic changes over the Holocene. Based on statistical analysis of proxy time series, Wanner el al., (2011) showed six specific cold events relapses in Northern Hemisphere during the Holocene (8.2 ka, 6.3 ka, 4.7 ka, 2.7 ka, 1.5 and 0.5 ka BP).

These episodes have alternately been attributed to freshwater forcing, volcanic activity, changes in Atlantic Meridional Overturning Circulation, and low solar variability (Wanner et al., 2011). Many studies have reported evidence for solar-forcing on Holocene climate change across a range of continental and marine archives (e.g., Neff et al., 2001; Bond et al., 2001; Soon et al., 2014; Munoz et al., 2015). Holocene solar activity variation showed several solar cycles on different temporal scales, Schwabe cycle (11 yr), the Gleissberg cycle (70-100 yr), Hale cycle (130 yr), de Vries cycle (200-210 yr), Eddy cycle (1000 yr), Bray or Halstatt cycle (2200-2400 yr). A number of climate proxies have been used to investigate solar forcing including geochemical and biological records from tree rings (Muraki et al., 2011), lake sediments (Kern et al., 2012), peatland (Turner et al., 2016) and speleothems (Niggemann et al., 2003).

During the last decades, speleothem studies have enhanced our knowledge of Holocene continental climate, increasing the chronological resolution and extending the period covered (Drysdale et al., 2009; Fleitmann et al., 2009; Genty et al., 2003; Wang et al. 2008). Their ability to be U-series dated with a precision of 0.5% (Edwards et al., 1987) combined with the possibility of layer counting when seasonal layers are present (Verheyden et al., 2006; Genty and Quinif, 1996), and even more if luminescent layering (Shopov et al., 1994) is present, makes speleothems a powerful tool to refine the chronology and increase the resolution of climate reconstructions. Speleothems contain several already well-studied climate proxies such as $\delta^{18}O$, $\delta^{13}C$, strontium isotopes and compositions (Fairchild and Baker, 2012; McDermott et al., 2011; Mangini et al., 2005; Mattey et al., 2008; Zhou et al., 2009; Goede et al., 1998). In addition, speleothem trace elements compositions represent a large proportion of speleothem proxies, used to reconstruct paleoenvironmental conditions. Variations in solar activity have been well documented in Holocene speleothems archives, by identifying changes in color or isotopic composition (e.g., Niggemann *et al*., 2003; Cosford et al*.,* 2009; Martín-Chivelet et al*.,* 2011) or variations in the thickness of speleothem layers (e.g., Genty and Quinif, 1996; Frisia et al*.,* 2003; Munoz et al*.,* 2009). As previously investigated in the Han-sur-Lesse cave from the same karstic system as Père Noël cave, and presented in Genty and Quinif (1996), the Han-stm-1 stalagmite has shown the presence of successive laminae forming very regular cycles of 11 years separated by a thick dark compact lamina, which is similar to the sunspot solar cycle. Until now, the potential of trace elements content in speleothems for recording solar activity variations is largely unexplored.

In previous low resolution studies, Verheyden et al. (2008, 2014) already observed that the PN stalagmite displays important variations in growth rate, geochemical (Mg/Ca and Sr/Ca) and isotopic (O, C) content along its longitudinal axis between 12.7 and 1.8 ka. Changes in isotopic equilibrium conditions were interpreted as due to changes in cave humidity and linked to changes in recharge, ie. precipitation minus evapo-transpiration. Changes in trace element content were interpreted as due to changes in water residence time linked to changes in water availability (Verheyden et al., 2008). In this study, by using high-resolution trace elements content (Ba, Sr, Mg and Al), we reconstruct climate changes that occurred in Northwestern Europe during the Holocene. We apply spectral and wavelet analysis of the trace elements time-series of the PN stalagmite to identify solar-type periodicities in the stalagmite record. Moreover, the sunspot reconstruction of Solanki et al., (2004) was compared with the PN times series to identify eventual solar cycles in the PN proxies and as such determine the possibilities for the study of solar activity changes over the Holocene.

2. **The state of the art of trace elements in speleothems**

To understand the significance of the elemental variations in speleothems (i.e., Ba, Sr, Al and Mg), it is necessary to investigate the potential sources of the chemical elements and their transfer through the karst system. Then, the relations between stable isotopes and trace elements are characterized and may be related to paleoclimate changes. Elemental trace contents may be derived from different sources such as natural and/or anthropogenic dust (local or distal) supplies (Allan et al., 2015; Dredge et al., 2013; Fairchild et al., 2000; Frisia et al., 2005; Wynn et al., 2008). They may also derive from the host rocks and adjacent soil cover (Tooth and Fairchild, 2003; Hartland et al., 2012; Treble et al., 2016). The trace elements could be transported along the soil profile by colloids or in dissolved forms depending on the element. The trace elements might show different forms of mobility (in dissolved and particulate form) and speed of migration in soils, depending on soil characteristics and processes (e.g., pH, organic acid content, redox conditions, leaching, ion exchange, temperature). Finally, the karst hydrology may also play a role in the speleothem trace element content (Fairchild et al., 2000; Tooth and Fairchild, 2003). Karstic processes include organic matter decomposition, dissolution, desorption and adsorption of colloids, Prior Calcite Precipitation (PCP), incorporation into carbonate minerals (e.g., Fairchild and Treble, 2009).

Previous studies have shown that the variations of trace element contents in a speleothem may provide information about changes in soil condition, water temperature, paleorainfall and hydrological conditions (e.g., Roberts et al., 1999, Fairchild et al., 2000; Fairchild and Treble, 2009). In temperate regions, trace element such as Ba, Sr and Mg tend to increase during drier periods when residence times are longer (Huang et al., 2001; Verheyden et al., 2000). Many studies performed on speleothems from different environmental settings reveal that both hydrological (e.g., amount of precipitation, groundwater residence time, rock-water interaction) and/or growth-related processes can affect trace element concentrations (e.g., Verheyden et al., 2000; Treble et al., 2003; Tremaine and Froelich, 2013). However, the link between trace element content in speleothems and climatic conditions can vary between caves settings due to differences in chemical composition of bedrock, groundwater movement, soil thickness and/or climatic conditions. Consequently, a thorough comparison between speleothem trace element content and another proxy (e.g., combining stable isotopes with trace elements and/or petrographic information and/or growth-rate) is important in order to minimize the uncertainties associated with stable isotope data interpretation (Fairchild et al., 2006; McDermott, 2004; and Verheyden et al., 2001).

3. Material and methods

PN stalagmite, 65 cm long, was taken in 2000 from the Belgian Père Noël cave (50.0°N, 5.2°E, 230 masl) in the Ardenne massif (Fig.1). The cave located about 200 km inland in a temperate maritime climate. The cave opened in the Devonian (Givetian) Fromelenne Formation with subvertical limestone beds (Delvaux de Fenffe, 1985) that reach a thickness of 70 m above the cave (Deflandre, 1986). The water entering the cave consists of local rain only, seeping directly through the overlying limestone. An approximately 40-cm-thick woodland soil covers the host rock limestone. A detailed description of climate, geology, and hydrological conditions were published in Verheyden et al. (2000, 2008, 2012).

The PN stalagmite was dated by TIMS U-series dating (9 dates) at the Department of Earth Sciences at Open University, UK, following the method described in McDermott et al. (1999). Calcite samples for U/Th dating (0.2–2 g) were spiked with a mixed ^{229}Th–^{236}U spike. U and Th were separated on columns loaded with 2 ml of anion exchange resin (AG1 X8, 200–400 mesh Biorad). The total procedural blanks were negligible (ca. 15 pg of Th and U). Errors

based on counting statistics were < 0.1‰ for all ratios except for $^{230}Th/^{229}Th$, where 2σ varied between 0.4 and 1.0% depending on the U content and age sample. Other dates (7 dates) were done by NEPTUNE Multi-Collector Inductively Coupled Plasma Mass Spectrometry (MC-ICP-MS) at the Laboratoire Géosciences Environnement Toulouse (GET) in France. U and Th were separated from the matrix and purified by ion exchange chromatography using AG1 X8 resin (Riotte and Chabaux, 1999; Violette et al., 2010). Errors based on counting statistics varied between 0.2 and 0.7 % for all ratios. Procedure blanks are <1 pg and ~10 pg for U and Th, respectively, which is negligible compared to the U and Th amounts in samples. A blank is measured between each sample and a standard (HU1) every four samples. All ages were corrected for detrital contamination assuming a $^{232}Th/^{238}U$ mass ratio of 3. One date was obtained in the Earth Science Department of the University of Minnesota on a Thermo-Scientific Neptune-Plus multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS). The procedures that were followed for uranium and thorium chemical separation and purification described in Edwards et al. (1987) and Cheng et al. (2009a, 2009b). Dating results are summarized in Table 1, with ages given in years before 1950. Errors given in the table (2σ) depend on the U and Th content and age of the sample.

The elementary geochemical composition (Ba, Sr, Mg and Al) of PN stalagmite was determined by Laser Ablation Inductively Coupled Plasma-Mass Spectrometer (LA-ICP-MS) mounted with an ESI New Wave UP-193FX Fast Excimer ArF laser of 193 nm at the Royal Museum for Central Africa (Tervuren, Belgium). Spots were made of 50 µm in diameter and spaced at 300-1000 µm intervals. The age model reveals a growth rate ranging between 0.02 and 0.65 mm.yr^{-1}. Consequently, the analysis of a diameter of 50 micrometers each 300-1000 micrometers interval corresponds to a sample of 1 to 3 years every 0.5 to 50 years. Detection limits are calculated from the intensity and standard deviation measurements of the blank. The limits of quantification range between 0.1µg g^{-1} for Sr, Al and Ba, and 2.5µg g^{-1} for Mg. Certified reference materials (NIST 610, NIST 612, NIST 614, MACS-1, and MACS-3) were analyzed with each series of samples, in order to determine the precision and accuracy of analytical procedures. Comparison between reference values and measured values were satisfactory (Table 2) within 65-98 %. For Ba, Sr, and Mg, the reproducibility was higher than 76%. The lowest value was observed for Al (65% for MACS-1 standard). The correlation coefficients (r) allow for the assessment of the degree of linear relationship between two variables. The correlation coefficient is significant when r values are close to the value 1 (0.5< r<1 : strong correlation, 0< r<0.5: low correlation).

To investigate the solar forcing controls on trace elements content in the PN stalagmite, we compared the concentrations of Ba, Sr, and Mg with sunspot number (solar activity-Solanki et al., 2004). Continuous wavelet transform was applied on the trace element time-series data to detect any significant periodicities. Prior to perform the spectral analysis each dataset was resampled as follows : for each age t (in BP) in the evenly spaced sequence 1789, 1799, 1809,..., the corresponding concentration of trace element is obtained through a linear interpolation between the two consecutive data points whose ages (in BP) surround t. Since 92% of the time gaps in the original time series are lower than 25 years and the remaining 8% all occur at once at the end of the signal (after 9.5 ka BP), the effects of such a resampling on the results presented are negligible (especially for periods such as the de Vries cycle or larger). More information in supplementary materials (Fig.6, 7, 8).

4. Results

The age model was constructed by using StalAge algorithm (Scholz and Hoffmann, 2011). The 17 ages obtained for stalagmite PN are all in stratigraphic order (Fig. 2). The age-depth model indicates that the PN stalagmite was deposited between 12.7 and 1.8 ka BP. The internal longitudinal section of the stalagmite presents a succession of brown parts and milky white parts along its longitudinal axis (Fig. 2). No clear hiatus is observed in the stalagmite. The longitudinal section also reveals variations in the stalagmite diameter along its longitudinal axis. The growth rate varied between 0.02 and 0.65 mm.yr^{-1} with the highest growth rates (0.09 to 0.65 mm.yr^{-1}) observed between 7.8 and 6.3 ka BP (i.e., between 37 and 12 cm). The lowest growth rates evidenced between 6.3 and 1.8 ka BP.

The Ba, Sr, Mg, and Al records in PN are composed of nearly six-hundred independent points. Profiles of Mg, Sr, Ba and Al in PN stalagmite are very similar (Fig. 3). The values (in µg g^{-1}) range from 5 to 65 for Ba, 20 to 160 for Sr, 1000 to 17600 for Mg and from 0.2 to 35 for Al. High trace elements contents are observed at different depths: from 540 to 455 mm (10.7-9.2 ka BP), 390 -370 mm (8.2-7.9 ka BP), between 300 and 140 mm (7.2-6.2 ka BP), and from 25 mm (3 ka BP) to the top of PN stalagmite at 2.02 ka BP (Fig.3, 4). In the low trace element content zones, the Al content was below the limit of detection (< 0.1 µg g^{-1}). Trace element results are closely correlated ($r_{(Sr, Ba)}$=0.77, $r_{(Mg, Sr)}$=0.72, $r_{(Mg, Ba)}$=0.48, , $r_{(Al, Ba)}$=0.42, $r_{(Al, Mg)}$=0.4g, Fig. 4), implying that similar processes influence their concentration changes, over all timescales. Strontium concentration averages approximately 2-4 times the Ba concentration, throughout the record. Trace element data was resampled at constant time interval to perform spectral analyses (more details in supplementary data). Significant periods of 71, 136, 198, 291, 497-602, 1029, 1365, 1706, 1861 and 2315 are observed in Ba time-series. Strontium also shows significant periodicities at 75, 133, 198, 281, 404-634, 960, 1280, 1365, 1855, and around 2630 yr. In addition, Mg data exhibit peaks around 68, 133, 209, 325, 500, 602-706, 967, 1533 and around 2365 yr. All observed periodicities display some similarities with solar variability, presented by sunspot number, which shows significant cycles at 68, 87, 136, 225, 352, 525, 975, 1462, 2275 yr (Fig.5) .

Stable isotopes data (δ^{18}O and δ^{13}C) from PN stalagmite reported in Verheyden et al. (2008). Stable isotope composition of PN varies between -4.3 and -6.4‰ for δ^{18}O (mean -5.4‰) and between -2.56 and -9.46‰ (VPDB) for δ^{13}C (mean -7.19 ‰) (Fig. 4). δ^{18}O displays a long term increasing trend from lower values (-6‰) between 12.7 and 10.7 ka BP to -5.3 ‰ between 10.7 and 7.5 ka BP and to higher values (>-5.0‰) between 6.3 and 1.8 ka BP. Lower values are observed at 10.7, between 9.5 and 9, between 8.3 and 6.2, at 5.5 ka BP and between 4.3 and 3.4 ka BP with a general increase until the end of speleothem deposition. The δ^{18}O and δ^{13}C values of PN stalagmite show very similar trends (Fig.4), confirmed by the rather high correlation coefficients of r =0.8. Consequently, we interpret the stable isotope signals recorded by PN stalagmite in terms of past climate variability.

5. Discussion

5.1. δ^{18}O, δ^{13}C and trace element

In previous studies (Verheyden et al., 2000, 2008; Genty and Deflandre, 1998), the PN stalagmite has received a great deal of scientific attention to 1) understand how climate signals are recorded in proxies speleothems of Père Noël cave (PN); 2) reconstruct climate variations at low resolution of the Holocene . The δ^{18}O and δ^{13}C of PN cave, has been suggested to reflect changes in the importance of evaporation and fast degassing, linked with changes in effective rainfall (Verheyden et al., 2008). To correctly interpret variations in δ^{18}O and δ^{13}C of PN speleothem, continuous monitoring of

12

several parameters was set up to investigate the controlling factors affecting the $\delta^{18}O$ and $\delta^{13}C$ of PN speleothem. Between 1991 and 1998, different cave parameters were measured such as cave air and seepage water temperature, humidity, P_{CO2}, drip water, $\delta^{18}O$ and $\delta^{13}C$ of seepage water, conductivity, trace element concentrations of the water and calcite. Several statements are derived from this monitoring:

(1) The mean annual precipitation of the nearby meteorological station (Lessive, located at 3 km NE of the cave), measured over the period 1980-1998, is 826 mm. The mean temperature inside the cave of 9 °C (varies between 8.5° and 9.2 °C) corresponds to the mean annual temperature of the air (9.2°C) at the nearby Lessive station.

(2) The relative humidity changes from 95 to 98%. There is a good correlation between the water excess (or rainfall), and the annual flowrate of the stalactite ($R^2= 0.98$). It demonstrates that the quantity of water feeding a stalagmite is controlled by the annual rainfall.

(3) the mean $\delta^{18}O$ of seepage water of -7.5‰ (-8.0 $<\delta^{18}O<$ -6.8 ‰) which is similar to annual mean rain water (-7.3‰). More negative $\delta^{18}O$ values of calcite occur during periods of higher cave effective precipitation when calcite deposition occurs closer to isotopic equilibrium.

(4) The stable isotopes signals ($\delta^{18}O$ and $\delta^{13}C$) were interpreted as due to variation in cave humidity and drip rate producing changes in the kinetics of the calcite deposition. Hendy tests indicated PN stalagmite was deposited out of equilibrium. The enrichment in both $\delta^{18}O$ and $\delta^{13}C$ in calcite and their positive and significant correlation are signs for a non-equilibrium precipitation.

(5) $\delta^{18}O$ and $\delta^{13}C$ of the stalagmite may give some information on dry/wet variations in the past: more negative $\delta^{18}O$ and $\delta^{13}C$ values occur during periods of higher cave water recharge, when calcite deposition occurs closer to isotopes equilibrium).

In this study, statistically significant positive correlations ($r_{(Sr, Ba)}=0.77$, $r_{(Mg, Sr)}=0.72$, $r_{(Mg, Ba)}=0.48$) are found between Sr, Ba and Mg over the Holocene period suggesting either a common or strongly related controlling process. Changes in element traces content in PN stalagmite are interpreted as due to changes in water residence time linked to changes in water availability. Higher Mg and Sr contents were previously observed during dry periods, characterized by longer water residence times (Verheyden et al., 2008) and associated to lower water availability. Since water residence time is related with water availability, trace elements in PN speleothem probably register changes in effective precipitation, i.e., precipitation minus evapotranspiration. The $\delta^{18}O$ and $\delta^{13}C$ and trace elements (Sr, Ba, and Mg) profiles show very similar trends, which is confirmed by the rather high correlation coefficients (r= 0.45-0.70), suggesting a common control. Previous studies (Verheyden et al., 2008) on PN stalagmite showed a similarity between $\delta^{18}O$ and $\delta^{13}C$ and Mg and Sr time-series, which may explain by two processes. First, Mg and Sr contents depend on their concentrations in the seepage waters and on the residence time of the water in the vadose zone, and consequently, through the recharge amount, on the rainfall amount. The dependence on the residence time might be due to the degree of "prior calcite precipitation" (PCP) that will increase both Mg and Sr in the seepage water and consequently in the calcite. $\delta^{18}O$ and $\delta^{13}C$ contents are controlled by the isotopic equilibrium state during precipitation of the calcite, which depends on the drip rate, and consequently, through recharge, on the rainfall amount too. Second, both chemical and isotopic compositions of the stalagmite depend on kinetic processes during calcite precipitation, which are linked with drip rate and water recharge. Consequently, chemical and isotopic ratios appear to be two proxies for the rainfall amount. In the PN speleothem, the

crossed link of stable isotopes ($\delta^{18}O$ and $\delta^{13}C$) as of trace element contents (Sr, Ba, and Mg) to water recharge may explain the covariation of the geochemical parameters over much of the Holocene period. In addition, this relationship may also be the consequence of kinetic effects during calcite precipitation as suggested by the observed similar variations of three parameters ($\delta^{18}O$ and $\delta^{13}C$ and trace elements) along a single layer of the Holocene stalagmite.

Aluminum can be used to determine the variations in the detrital (non-carbonate) content in speleothems. These particles may be transported attached to particles and colloids during periods of intense precipitation, which results in high drip rates (White, 1997; Schimpf et al., 2011). In PN stalagmite, in the low trace element content zones, the Al content was below the limit of detection (< 0.1 µg g^{-1}) (from 10.2 to 9.3 ka, between 8.1 and 7.2 ka and from 4.1 to 2.8 ka BP; Fig.3, 4). However, Al concentration was higher than the detection limit during the cold events and its maximum values coincide with maxima in the other investigated trace elements (Fig.4). High Al content reflecting a high content of detrital material (particles/colloids) would suggest higher flow rates in the aquifer and thus wetter conditions. In contrast high values of Sr, Ba and Mg reflect drier conditions. This contradiction can be explain as follows as: during the drier periods trace elements (Sr, Ba, and Mg) continue to accumulate in the vadose zone, where the residence time is long, then a high flow rate flashed trace elements and leaded them to precipitate in the cave. During wetter periods, the high drip rates may be transport Al "attached to particles and colloids" from the soil and the trace elements from vadose zone to the cave environment.

5.2. **Spectral and wavelet analyses**

First spectral analysis demonstrates that trace element time-series contain dominant periodicities on decadal to millennial times scales (Fig.5). The most frequently occurring periodicities were those at 68-75, 133-136, 198-209, 291-358, 404-602,912-1029 and 2365-2670 yr (Fig.5). Those periodicities are confirmed by wavelet results (Fig. 6). Second we test PN time-series for solar forcing by applying wavelet approach. Solar forcing was represented by the number of sunspots (Solanki et al., 2004). Wavelet analyses of Ba, Sr and Mg time-series with reconstructed sunspot number time series reveal significant common periodicities within (63–80, 133-140, 198–220, 514-561, 912-1029 and 2245-2670 yr-Fig.6, supplementary data). Those periodicities coincide with known solar cycles (e.g., Steinhilber et al., 2012; McCracken et al., 2013) and are in agreement with decadal-centennial scale variability in Holocene climatic records from widely dispersed geographic regions. For example, the 63–80 yr interval is similar to the Gleissberg cycle (70-100 yr) whereas the 133-140 yr frequency interval corresponds to the Hale cycle (130 yr). The 198–220 yr cycle of PN record is close to the de Vries (200-210 yr) solar cycle (Steinhilber et al., 2012). De Varies's cycle was already observed in numerous palaeoclimate records all over the world (Wanger et al., 2001; Duan et al., 2014; Czymzik et al., 2016). There are also strong variability around 281-325 yr and 535 yr (514-561) periods that correspond to solar variability that reported as "350 and 500 unnamed cycles" (Steinhilber et al., 2012). These cycles were also detected in numerous other palaeoclimatic records worldwide (e.g., Soon et al., 2014; Chapman and Shackleton, 2000). The 960-1029 yr cycle of the speleothem could match the Eddy cycle (1000 yr) as recognized in both ice cores (Stuiver et al., 1995) and marine sediments (e.g., Chapman and Shackleton, 2000). The trace elements PN records show small peaks between 1280 and 1533 yr (Fig. 5). This periodicity is identical to the Bond cycle (1470 ± 500 yrs) detected from ice rafted debris in North Atlantic sediment cores (Bond et al., 2001). Finally the 2245-2670 yr cycle, could be related to the Bray or Halstatt cycle (2200-2400 yr) that is recognized in other palaeoclimatic records (e.g., kern et al., 2012). The significant common periodicities between trace elements PN records and Solar records suggest that solar variability

influenced PN trace elements content on decadal to millennial time scales. Since the changes of trace elements in the PN speleothem were formerly demonstrated as due to changes in recharge, i.e. effective precipitation, the study suggests that variations in solar activity may be a significant forcing on either the precipitation or on the soil activity. Climate models and paleoclimate proxies (Shindell et al., 2001; Kodera, 2002; Rimbu et al., 2004) have implicated North Atlantic Oscillation (NAO) variability as an important element in the climate response to variable solar forcing. The NAO is known to influence precipitation $\delta^{18}O$ ($\delta^{18}O_p$) through its control on air temperature and on the trajectory of the westerly winds that carry moisture onto Europe during boreal winters (Smith et al., 2016). During the periods of low solar activity, NAO negative conditions caused a southward displacement of moisture delivery to Europe, and are associated with increased humidity in Central and Southern Europe (Smith et al., 2016; Magny, 2004). However, this does not necessarily imply that solar forcing was the main source of all Holocene climate variability since different dynamical processes, such as explosive volcanic eruptions, fluctuations of the ocean thermohaline circulation or internal feedbacks, might also have played an important role (Wanner et al., 2008).

5.3. **Relation between trace element records and known Holocene climate events**

The Holocene records PN indicate that the maxima of trace elements concentrations coincide with the dry/cold events (Fig. 7) defined in Wanner et al. (2011), which partly coincide with the events of Bond et al. (2001). The higher positive correlations between trace elements concentrations are observed during 4 cold periods (i.e., from 10.7 to 10.3 ka BP, $r_{(Sr, Ba)}$=0.96, $r_{(Mg, Sr)}$=0.49, $r_{(Mg, Ba)}$=0.45; from 9.5 to 9.3 ka BP, $r_{(Sr, Ba)}$=0.94, $r_{(Mg, Sr)}$=0.80, $r_{(Mg, Ba)}$=0.70; around 8.2 ka, , $r_{(Sr, Ba)}$=0.91, $r_{(Mg, Sr)}$=0.63, $r_{(Mg, Ba)}$=0.47; from 6.4 to 6.2 ka BP, $r_{(Sr, Ba)}$=0.95, $r_{(Mg, Sr)}$=0.60, $r_{(Mg, Ba)}$=0.42; from 4.8 to 4.6 ka BP, $r_{(Sr, Ba)}$=0.98, $r_{(Mg, Sr)}$=0.97, $r_{(Mg, Ba)}$=0.92; around 2.7 ka BP, $r_{(Sr, Ba)}$=0.98, $r_{(Mg, Sr)}$=0.70, $r_{(Mg, Ba)}$=0.70). In marine sediments Bond et al. (1997, 2001) revealed 1470 yrs± 500 Holocene cycles. Nine Bond cycles were detected over the Holocene and peaked around 0.4, 1.4, 2.8, 4.3, 5.9, 8.1, 9.4, 10.3 and 11.1 ka BP (Bond et al., 1997, 2001; Wanner and Bütikofer, 2008b). These events, attributed to lower solar activity, most probably triggered the slowdown of the thermohaline ocean circulation over the North Atlantic and European regions (Renssen et al., 2007). Figure 7 shows a comparison of the trace elements concentrations in the PN stalagmite and the frequency of warm/cold and wet/dry events in the Northern Hemisphere (Wanner et al., 2014). In the PN stalagmite, we identify several periods of dry climate, these are centered at 10.5, 9.3, 8.2, 6.3, 5.4, 4.6 and around 2.7 ka BP, and their duration ranges from 100 to 400 yr (Fig. 7). These intervals characterized by higher trace elements contents in PN record were interpreted as dryer periods.

5.3.1.   Younger Dryas and Early Holocene

The PN stalagmite starts its growth on the underlying limestone block at 12.7 ka BP, indicating favorable climatic and environmental conditions for stalagmite growth. The larger diameter of the stalagmite, at ~12.0 ka BP, compared to its mean diameter suggests that there is enough water to flow down on the flank of the stalagmite and to precipitate calcite. The inception of speleothem growth is in agreement with the beginning of the cold Younger Dryas (YD) event (12.9-11.7 ka BP). In addition, the decreasing $\delta^{13}C$ values indicate a progressively increasing soil activity. The still rather low $\delta^{18}O$ (Fig.4) suggest relatively wet conditions in agreement with the important stalagmite diameter. This is synchronous with the warm climate during the early YD (12.9–12.15 ka BP) found in European lake sediments from Northern Spain, Norway and Western Germany (Baldini et al., 2015; Bakke et al., 2009; Brauer et al., 2008). The high-resolution records from those three lakes suggested that warming climate associated with the early YD may be related

to local climate change (Lane et al., 2013; Baldini et al., 2015). Our high-resolution trace elements from PN stalagmite consolidate the concept that the early YD (12.9–12.15 ka BP) was humid and/or warm in SW and NW Europe. The period, from 12.1 to about 11.7 ka BP, is characterized by a gradual increase of trace element concentrations, suggesting a climate change from wet to dry conditions (Fig.4, 7). The $\delta^{18}O$ values increase from -6.3 to -5.5 ‰ suggests relatively dry conditions in agreement with trace element concentrations (Fig.4). European archives (lake and speleothem records) indicate that a cooler and/or drier climate characterized the YD termination (12.1-11.7 ka BP) (e.g., Genty et al., 2006; Von Grafenstein et al., 1999). This rapid climate shift attests for the major influence of the North Atlantic Ocean circulation on the YD/Holocene climate transition (Pearce et al., 2013; Baldini et al., 2015).

During the Early Holocene, i.e. before 7 ka BP, the amplitude of the frequency of warm/cold or dry/ wet events was from the rapidly melting ice sheets of the Northern Hemisphere (Carlson et al., 2014). The Holocene started about 11.7 ka BP with a transition from a cool YD to a wet and/or warm climate state. In the PN stalagmite, low trace element contents suggest that wet conditions persisted at the beginning of the Holocene (Fig.4). The $\delta^{18}O$ and $\delta^{13}C$ values decrease from -5.5 to -6.1 ‰ and -6.3 to -8.5 ‰ (respectively) suggests relatively wet conditions in agreement with trace elements concentrations. PN speleothem suggests a wet Early Holocene with dryer conditions from 10.7 to 10.3, at 10.0, at 9.7, at 9.2 and from 8.5 to 8.2 ka BP (Verheyden et al., 2014). Trace element contents in the PN stalagmite display significant variability during the Early Holocene, with three maxima observed around 10.5, from 9.5 to 9.2 and around 8.2 ka BP (Fig. 4, 7). The covariance between geochemical proxies support, as explained in Verheyden et al. (2008), their interpretation in terms of dry-wet changes with higher trace elements and higher $\delta^{18}O$ values linked to drier conditions. The drying is in agreement with a decrease in speleothem growth rate and diameter. Moreover, a denser calcite is deposited at 10.4 ka, between 9. **5**and 9.2 and between 8.4 and 8.2 ka BP (Fig.4). The highest concentrations of trace elements in PN correspond to low sunspot numbers and to cold periods (Fig. 4), as defined in Bond et al., (2001). During the three intervals (i.e., around 10.5, between 9. **5**and 9.2 and between 8.4 and 8.2 ka BP), the wavelet spectrum of trace elements shows that the solar imprint is well present in the PN stalagmite (Fig.6). Those three intervals may correspond to Bond cycles that caused by the lower solar activity additionally triggered slowdown of the thermohaline ocean circulation in the North Atlantic and European regions (Renssen et al., 2007). Continuous cyclic periods of around 500 and 1000 yrs are presented, even with different spectral powers for the different elements, during the interval 10.7-10.3 ka BP. The 1000-yr band is especially pronounced for Ba and Sr than Mg. For the intervals from 9. **5**to 9.2 and between 8.4 and 8.2 ka BP, continuous cyclic periods of around 80, 130, 500, 1000, 1300-1500 yr are present. Ice, speleothems and sediment records from the North Atlantic region demonstrate that the Early Holocene period was relatively stable and interrupted by periods of short-lived cooling such as: 10.3 ka (Bjorck et al., 2001), 9.3 ka (e.g., Boch et al., 2009; Rasmussen et al., 2007; von Grafenstein et al., 1999) and 8.2 ka event (e.g., Alley et al., 1997; Ellison et al., 2006). Solar variability and/or changes in Atlantic Meridional Overturning Circulation have been proposed as causes of climate changes in the Early Holocene (Björck et al., 2001; Marshall et al., 2007; Plunkett and Swindles, 2008). A cold event around 8.2 ka BP has been recorded in the North Atlantic and Northern Europe (Alley et al., 1997; Barber et al., 1999; Clark et al., 2001). This event has been well recorded in PN stalagmite and discussed in detail in Verheyden et al., (2014). The correlation between trace elements and sunspot number implies that periods of lower intensity of solar irradiation were probably accompanied by drier climate in NW Europe. However, solar forcing cannot alone explain the variability of the Early Holocene signal. Previous studies in ice cores (e.g. Alley et al., 1997) or lake sediments (e.g.,

Prasad et al., 2006), demonstrated that the climate changes in Western Europe and Greenland are similar during the Early Holocene and may be caused by changes in solar activity, and changes in North Atlantic thermohaline circulation.

5.3.2. Mid and Late Holocene

The interval from 8.1 to about 7 ka BP is characterized by low contents of trace elements and low $\delta^{18}O$ and $\delta^{13}C$ values suggesting relatively wet conditions. This phase coincides with the interval of maximum frequency period of warm and wet events detected in the Northern Hemisphere (Wanner et al., 2014) (Fig.7). The interval between about 7 and 4.2 ka BP "called Holocene Climate Optimum" is characterized by higher summer temperatures in the Northern Hemisphere (e.g., Alverson et al., 2003). High trace elements values were detected in PN stalagmite during three intervals, from 6.4 to 6.2, 5.5 to 5.4 and from 4.7 to 4.5 ka BP (Fig.4). The period between 7.3 and 6.0 ka BP corresponds to the warmest period of the Holocene in Greenland (Jonhsen et al., 2001). From 6.4 to 6.2 ka BP a high positive correlation (r=0.45-0.95) was observed between element trace concentrations confirming dryer conditions. The drying is in agreement with a decrease in the speleothem growth rate and diameter. The high concentrations of trace elements in PN correspond to low sunspot number (Fig. 4). This phase coincides with the period of maximum frequency of dry events detected in the Northern Hemisphere (Wanner et al., 2014) and with a cold event as defined in Wanner et al., (2011). Between 5.5 and 5.4 ka BP, the trace elements (high values) display an anticorrelation with sunspot number (low values) suggesting relatively dry conditions (Fig.4). During this period, trace elements present frequencies corresponding to the Gleissberg cycle (70-100 yr), Hale cycle (130 yr), de Vries cycle (200-210 yr), 500 unnamed cycle and Eddy cycles (1000 yr) (Fig.6).

High trace elements and $\delta^{18}O$ values were detected in the PN stalagmite from 4.7 to 4.5 ka BP corresponding to a dry period. The high positive correlation (r=0.78-0.97) observed between trace element concentrations during this period confirms dry conditions. The period between 4.7 and 4.5 ka BP corresponds to the cold event (4.6-4.8) defined by Wanner et al., (2011) and coincides roughly with Bond events. This phase coincides with the period of decrease in frequency of wet events and increased of dry events detected in the Northern Hemisphere (Fig.7). In addition, our spectral analysis shows the prominence of 63–80, 133-140, 198–220, 514-561, and 912-1029 year periodicities in the PN stalagmite (Fig.6).The periodicities found here may suggests evidence for solar-forced climate change because they match the ranges of cycles in solar reconstructions. From 4.5 to 3.1 ka BP, general climatic conditions favor the speleothem deposition. Low values for trace elements and for $\delta^{18}O$ and $\delta^{13}C$ indicate relatively wet and temperate conditions.

General dry conditions as showed by PN proxies were observed from 3 ka BP to the end of the speleothem deposition at 1.8 ka. The final dry phase may be related to the cool event showed by Wanner et al. (2011) that occurred between 3.3 and 2.5 ka BP and corresponds with Bond event (Fig. 4).This dry period also coincides with low solar activity (Fig. 4). This period was caused by an abrupt decrease of solar activity and has been reported from different proxies in Greenland and Europe (e.g., van Geel et al., 1996; Blaauw et al., 2004; Plunkett and Swindles, 2008; O'Brien et al, 1995). The overall good agreement of the timing of the dry/wet or cold phases recorded in the PN stalagmite with the timing of the Wanner and Bond events (Wanner et al., 2001, 2014; Bond et al., 1997, 2001) confirms the potential of the PN speleothem to reconstruct the Holocene paleoclimate.

6. Conclusion

We have shown that the high-resolution trace element records obtained by LA-ICP-MS from the PN stalagmite provide a detailed paleoclimate and/or paleoenvironment record of Northwestern Europe through the Holocene. The strong covariation of trace elements (Ba, Sr, and Mg) with $\delta^{18}O$ and $\delta^{13}C$, confirms a common or strongly related controlling process. Based on trace element time-series we demonstrate that several events at 10.3, 9.3-9.5, around 8.2, 6.4-6.2, 4.7-4.5, and around 2.7 ka BP alternate with periods of relatively stable and dry climate. These intervals coincide with the cold events defined in marine and continental archives. The trace element time-series of the PN speleothem reveals a significant correlation with sunspot number records, suggesting some solar forcing in the PN trace elemental records. This observation is confirmed by wavelet analyses that reveal common solar periodicities (Gleissberg cycle, de Vries cycle, unnamed 500 year, Eddy cycles, and Hallstatt cycle) in agreement with those recognized in the North Atlantic marine cores and the Greenland ice cores, as well as some other terrestrial Holocene records, suggesting common forcing mechanisms. Our study, based on high-resolution LA-ICP-MS analyses, emphasizes that speleothem trace element profiles may be considered as a new solar activity proxy on decadal-centennial timescales over the Holocene.

Reference

Allan, M., Fagel, N., Van Rampelbergh, M., Baldini, J., Riotte, J., Cheng, H., Edwards, R.L., Gillikin, D., Quinif, Y., Verheyden, S.: Lead concentrations and isotope ratios in speleothems as proxies for atmospheric metal pollution since the industrial revolution. Chem. Geol. 401, 140–150, 2015.

Allan, M., Le Roux, G., Piotrowska, N., Beghin, J., Javaux, E., Court-Picon, M., Mattielli, N., Verheyden S., and Fagel, N.: Mid and late Holocene dust deposition in Western Europe: The Misten peat bog (Hautes Fagnes - Belgium). Clim. Past, 9, 2285-2298, 2013.

Alley, R.B., Mayewski, P.A., Sowers, T., Stuiver, M., Taylor, K.C., Clark, P.U.: Holocene climatic instability: A prominent, widespread event 8200 yr ago. Geology, 25, 483 – 486, 1997.

Alverson, K.D., Bradley, R.S., Pedersen, T.F.: Paleoclimate, Global Change and the Future. Springer, New York, 2003.

Baldini, L.M., McDermott, F., Baldini, J.U.L., Arias, P., Cueto, M., Fairchild, I.J., Hoffmann, D.L., Mattey, D.P., Müller, W., Nita, D.C., Ontañón, R., Garciá-Moncó, C., Richards, D.A.: Regional temperature, atmospheric circulation, and sea-ice variability within the Younger Dryas Event constrained using a speleothem from northern Iberia. Earth and Planetary Science Letters 419, 101- 110, 2015.

Barber, D. C., Dyke, A., Hillaire-Marcel, C., Jennings, A. E., Andrews, J. T., Kerwin, M. W., Bilodeau, G., McNeely, R., Southon, J., Morehead, M. D., and Gagnon, J. M.: Forcing of the cold event 8200 years ago by catastrophic drainage of laurentide lakes, Nature, 400, 5 344–348, 1999.

Bakke, J., Lie, O., Heegaard, E., Dokken, T., Haug, G.H., Birks, H.H., Dulski, P., and Nilsen, T.: Rapid oceanic and atmospheric changes during the Younger Dryas cold period. Nature Geoscience, 2, 202–205, 2009.

Bjorck, S., Muscheler, R., Kromer, B., Andresen, C.S., Heinemeier, J., Johnsen, J., Johnsen, S.J., Conley, D., Koc, N., Spurk, M., Veski, S.: Highresolution analyses of an early Holocene climate event may imply decreased solar forcing as an important climate trigger. Geology, 29, 1107–1110, 2001.

Blaauw, M., van Geel, B. and van der Plicht, J.: Solar forcing of climate change during the mid-Holocene: indications from raised bogs in The Netherlands. The Holocene 14, 35-44, 2004.

Boch, R., Spotl, C., and Kramers, J.: High-resolution isotope ¨ records of early Holocene rapid climate change from two coeval stalagmites of Katerloch Cave, Austria, Quaternary Sci. Rev., 28, 2527–2538, 2009.

Bond, G., Showers, W., Cheseby, M., Lotti, R., Alsami, P., deMenocal, P., Priore, P., Cullen, H., Hajdas, I., and Bonani, G.: A pervasive millennial-scale cycle in North Atlantic Holocene glacial climates, Science, 278, 1257–1266, 1997.

Bond, G., Kromer, B., Beer, J., Muscheler, R., Evans, M.N., Showers, W., Hoffmann, S., LottiBond, R., Hajdas, I., Bonani, G.: Persistent solar influence on North Atlantic climate during the Holocene. Science 294, 2130–2136, 2001.

Brauer, A., Haug, G.H., Dulski, P., Sigman, D.M., Negendank, J.F.W.: An abrupt wind shift in western Europe at the onset of the Younger Dryas cold period. Nature Geoscience 1, 520-523, 2008.

Chapman, M. R. and Shackleton, N.: Evidence of 550-year and 1500-year cyclicities in North Atlantic circulation pattern during the Holocene, The Holocene, 10(3), 287–291, 2000.

Cheng, H., Edwards, R.L., Broecker, W.S., Denton, G.H., Kong, X., Wang, Y., Zhang, R.,Wang, X.: Ice Age terminations. Science, 326, 248-252,2009a.

Cheng, H., Fleitmann, D., Edwards, R.L., Wang, X.F., Cruz, F.W., Auler, A.S.,Mangini, A., Wang, Y.J., Kong, X.G., Burns, S.J., Matter, A.: Timing andstructure of the 8.2 kyr BP event inferred from delta O-18 records of stalagmitesfrom China, Oman, and Brazil. Geology , 37, 1007-1010, 2009b.

Carlson, A. E., Winsor, K., Ullman, D. J., Brook, E., Rood, D. H., Axford, Y., Le Grande, A. N., Anslow, F., and Sinclair, G.: Earliest Holocene south Greenland ice-sheet retreat within its late-Holocene extent, Geophys. Res. Lett., 41, 5514–5521, 2014.

Clark, P.U., Marshall, S.J., Clarke, G.K.C., Hostetler, S.W., Licciardi, J.M., Teller, J.T.: Freshwater forcing of abrupt climate change during the last glaciation. Science, 293, 283–287, 2001.

Cosford, J., Qing, H., Mattey, D., Eglington, B. and Zhang, M.: Climatic and local effects on stalagmite δ13C values at Lianhua Cave, China. Palaeogeog. Palaeoclimat. Palaeoecol., 280, 235-244, 2009.

Czymzik, M., Muscheler, R., Brauer, A.: Solar modulation of flood frequency in central Europe during spring and summer on interannual to multi-centennial timescales. Climate of the Past, 12(3), 799-805, 2016.

Deflandre, G. : La grotte du Pere Noel. De la Meuse a l'Ardenne 2, 5–56, 1986.

Delvaux de Fenffe, D. : Geologie et Tectonique du parc de Lesse et Lomme au bord sud du Bassin de Dinant (Rochefort, Belgique). Bull. Soc. Belge Geol., 94(1) , 81–95, 1985.

Delmonte, B., Petit, J. R., and Maggi, V.: Glacial to Holocene implications of the new 27,000-year dust record from the EPICA Dome C (East Antarctica) ice core, Clim. Dynam., 18, 647–660, 2002.

Dredge, J., Fairchild, I.J., Harrison, R.M., Fernandez-Cortes, A., Sanchez-Moral, S., Jurado, V., Gunn, J., Smith, A., Spoetl, C., Mattey, D., Wynn, P.M., Nathalie, G.: Cave aerosols: distribution and contribution to speleothem geochemistry. Quatern. Sci. Rev. 63, 23–41, 2013.

Drysdale, R.N., Hellstrom, J.C., Zanchetta, G., Fallick, A.E., Sanchez Go, M.F., Couchoud, I., McDonald, J., Maas, R., Lohmann, G., Isola, I.: Evidence for obliquity forcing of glacial termination II. Science 325, 1527–1531, 2009.

Duan, F.C., Wang, Y.J., Shen, C.-C., Wang, Y., Cheng, H., Wu, C.-C., Hu, H.-M., Kong, X.G., Liu, D.B., Zhao, K.: Evidence for solar cycles in a late Holocene speleothem record from Dongge Cave, China. Scientific Reports 4, 5159, 2014.

Edwards, R.L., Chen J.H., Wasserburg, G.J.: 238U-234U-230Th-232Th systematics and the precise measurement of time over the past 500.000 years, Earth Plan. Sci. Lett., 81, 175-192, 1987.

Ellison, C.R.W., Chapman, M.R., Hall, I.R.: Surface and deep ocean interactions during the cold climate event 8200 years ago. Science, 312, 1929 – 1932, 2006.

Fairchild, I.J., Borsato, A., Tooth, A.F., Frisia, S., Hawkesworth, C.J., Huang, Y., McDermott, F., Spiro, B.: Controls on trace element (Sr–Mg) compositions of carbonate cave waters: implications for speleothem climatic records. Chem. Geol, 166, 255–269, 2000.

Fairchild, I. J., Smith, C. L., Baker, A., Fuller, L., Spotl, C., Mat- ¨ tey, D., McDermott, F., and E.I.M.F.: Modification and preservation of environmental signals in speleothems, Earth Sci. Rev., 75, 105–153, 2006.

Fairchild, I. J. and Treble, P. C.: Trace elements in speleothems as recorders of environmetal change, Quaternary Sci. Rev., 28, 449– 468, 2009.

Fairchild, I. J. and Baker, A.: Speleothem Science: from process to past environments, Wiley Blackwell, ISBN: 978-1-4051-9620-8, 448 pp., 2012.

Fleitmann, D., Cheng, H., Badertscher, S., Edwards, R.L., Mudelsee, M., Gokturk, O.M., Fankhauser, A., Pickering, R., Raible, C.C., Matter, A., Kramers, J., Tüysuz, O.: Timing and climatic impact of Greenland interstadials recorded in stalagmites from northern Turkey. Geophys. Res. Lett. 36. doi:10.1029/ 2009GL0, 2009.

Frisia, S., Borsato, A., Preto, N. and McDermott, F.: Late Holocene annual growth in three Alpine stalagmites records the influence of solar activity and the North Atlantic Oscillation on winter climate: Earth Planet. Sci. Lett., 290 216, 411-424, 2003.

Frisia, S., Borsato, A., Fairchild, I. J., and Susini, J.: Variations in atmospheric sulphate recorded in stalagmites by synchrotron micro-XRF and XANES analyses, Earth Planet. Sci. Lett., 235, 729–740, 2005.

Gabrielli, P., Wegner, A., Petit, J.R., Delmonte, B., De Deckker, P., Gaspari, V., Fischer, H., Ruth, U., Kriews, M., Boutron, C., Cescon, P., and Barbante, C.: A major glacial-interglacial change in aeolian dust composition inferred from Rare Earth Elements in Antarctic ice, Quaternary Sci. Rev., 29, 265–273, 2010.

Genty, D., Blamart, D., Ghaleb, B., Plagnes, V., Causse, Ch., Bakalowicz, M., Zouari, K., Chkir, N., Hellstrom, J., Wainer, K., Bourges, F.: Timing and dynamics of the last deglaciation from European and North African δ 13C stalagmite profiles — comparison with Chinese and South Hemisphere stalagmites. Quat. Sci. Rev. 25, 2118–2142, 2006.

Genty, D., Blamart, D., Ouahdi, R., Gilmour, M., Baker, A., Jouzel, J., and Van-Exter, S.: Precise dating of Dansgaard-Oeschger climate oscillations in western Europe from stalagmite data, Nature, 421, 833-837, 2003.

Genty, D. and Quinif, Y.: Annually laminated sequences in the internal structure of some Belgian stalagmites - Importance for paleoclimatology, Journal of Sedimentary Research, 66, 275-288, 1996.

Goede, A., McCulloch, M., McDermott, F., Hawkesworth, Ch.: Aeolian contribution to strontium and strontium isotope variations in a Tasmanian speleothem. Chem. Geol., 149, 37–50, 1998.

Hartland, A., Fairchild, I.J., Lead, J.R., Borsato, A., Baker, A., Frisia, S., Baalousha, M.: From soil to cave: transport of trace metals by natural organic matter in dripwaters. Chem. Geol., 304, 68–82, 2012.

Huang, Y., Fairchild, I.J.: Partitioning of Sr2+ and Mg2+ into calcite under karstanalogue experimental conditions. Geochim. Cosmochim. Acta, 65 (1), 4762, 2001.

Johnsen, S. J., Dahl-Jensen, D., Gundestrup, N., Steffensen, J. P., Clausen, H. B., Miller, H., Masson-Delmotte, V., Sveinbjörnsdottir, A. E., and White, J.: Oxygen isotope and palaeotemperature records from six Greenland ice-core stations: Camp Century, Dye-3, GRIP, GISP2, Renland and NorthGRIP, J. Quaternary Sci., 16, 299–307, 2001.

Kern, A. K., Harzhauser, M., Piller, W. E., Mandic, O., and Soliman, A.: Strong evidence for the influence of solar cycles on a Late Miocene Lake system revealed by biotic and abiotic proxies, Palaeogeogr. Palaeocl., 329–330, 124–136, 2012.

Kodera, K.: Solar cycle modulation of the North Atlantic Oscillation: Implication in the spatial structure of the NAO. Geoph. Res. Let., 29 (8), 2002

Lane, C.S., Brauer, A., Blockley, S.P.E., Dulski, P.: Volcanic ash reveals time-transgressive abrupt climate change during the Younger Dryas. Geology ,41, 1251-1254, 2013.

Martín-Chivelet, J., Muñoz-García, M.B., Edwards, R.L., Turrero, M.J. and Ortega, A.I.: Land surface temperature changes in Northern Iberia since 4000yr BP, based on δ13C of speleothems. Global Planet. Change, 77, 1-12, 2011.

Magny, M.: Holocene climatic variability as reflected by midEuropean lake-level fluctuations, and its probable impact on pre-historic human settlements, Quaternary Int., 113, 65–80, 2004.

Mangini A., Spotl C. and Verdes P. (2005) Reconstruction of temperature in the Central Alps during the past 2000 yr from a d18O stalagmite record. Earth Planet. Sci. Lett. 235, 741–75, 2005.

Marx, S. K., McGowan, H. A., and Kamber, B. S.: Long-range dust transport from eastern Australia: a proxy for Holocene aridity and ENSO-induced climate variability, Earth Planet. Sci. Lett., 282, 167–177, 2009.

Marx, S. K., Kamber, B. S., McGowan, H. A., and Denholm, J.: Holocene dust deposition rates in Australia's Murray-Darling Basin, record the interplay between aridity, position of the mid-latitude westerlies, Quaternary Sci. Rev., 30, 3290–3305, 2011.

Mattey, D., Lowry D., Duffet, J., Fisher, R., Hodge, E., and Frisia, S.: A 53 year seasonally resolved oxygen and carbon isotope record from a modern Gibraltar speleothem: reconstructed drip water and relationship to local precipitations. Earth Plant. Sci. Lett., 269, 80–95, 2008.

Mayewski, P. A., Rohling, E. E., Stager, J. C., Karlen, W., Maasch, K. A., Meeker, L. D., Meyerson, E. A., Gasse, F., van Kreveld, S., Holmgren, K., Lee-Thorp, J., Rosqvist, G. Rack, F., Staubwasser, M., Schneider, R. R., and Steig, E. J.: Holocene climate variability, Quaternary Res., 62, 243–255, 2004.

Marshall, J.D., Lang, B., Crowely S.F., Graham, P.W., Van Clasteren, P., Fischer, E.H., et al.: Terrestrial impact of abrupt changes in the North Atlantic thermohaline circulation: Early Holocene, UK. Geology, 35, 639 – 642, 2007.

McDermott, F., Atkinson, T.C., Fairchild, I.J., Baldini, L.M. and Mattey, D.P.: A first evaluation of the spatial gradients in δ18O recorded by European Holocene speleothems. Global Plan. Change, 79, 275-287, 2011.

McDermott, F.: Palaeo-climate reconstruction from stable isotope variations in speleothems: a review. Quaternary Sci. Rev., 23, 901-918, 2004.

McCracken, K.G., Beer, J., Steinhilber, F., Abreu, J.: A phenomenological study of the cosmic ray variations over the past 9400 years, and their implications regarding solar activity and the solar dynamo. Solar Physics 286, 609–627, 2013.

Munoz, A., Bartolome, M., Munoz, A., Sancho, C., Moreno, A., Hellstrom, J.C., Oscan, M.C., Cacho, I.: Solar influence and hydrological variability during the Holocene from a speleothem annual record (Molinos Cave, NE Spain). Terra Nova, 27, 300–311, 2015.

Muraki, Y., Masuda, K., Nagaya, K., Wada, K., Miyahara, H.: Solar variability and width of tree ring. Astrophys Space Sci Trans, 7, 395–401, 2011.

Neff, U., Burns, S.J., Mangini, A., Mudelsee, M., Fleitmann, D. and Matter, A.: Strong coherence between solar variability and the monsoon in Oman between 9 and 6 kyr ago. Nature 411, 290 – 293, 2001.

Niggemann, S., Mangini, A., Mudelsee, M., Richter, D.K., and Wurth, G.: Sub-Milankovitch climatic cycles in Holocene stalagmites from Sauerland, Germany: Earth Planet. Sci. Lett., 216, 539–547, 2003.

O'Brien, S.R., Mayewski, P.A., Meeker, L.D., Meese, D.A., Twickler, M.S., Whitlow, S.I.: Complexity of Holocene climate as reconstructed from a Greenland ice core. Science, 270, 1962–1964, 1995.

Pearce, C., Seidenkrantz, M.-S., Kuijpers, A., Massé, G., Reynisson, N.F., and Kristiansen, S.M.: Ocean lead at the termination of the Younger Dryas cold spell. Nature Communications, 4, 1664, 2013.

Plunkett, G., and Swindles, G.T.: Determining the Sun's influence on Late Glacial and Holocene climates: a focus on climate response to centennial-scale solar forcing at 2800 cal. BP. Quarter. Sci. Rev., 27, 175–84, 2008.

Prasad, S., Brauer, A., Rein, B. & Negendank, J. F. W.: Rapid climate change during the early Holocene in western Europe and Greenland. The Holocene, 16, 153-158, 2006.

Rasmussen, S. O., Vinther, B. M., Clausen, H. B., and Andersen, K. K.: Early Holocene climate oscillations recorded in three Greenland ice cores, Quat. Sci. Rev., 26, 1907–1914, 2007.

Renssen, H., Goosse, H., Fichefet, T.: Simulation of Holocene cooling events in a coupled climate model. *Quat. Sci. Rev.*, 26, 2019–2026, 2007.

Roberts, M., Smart, P., Hawkesworth, C., Perkins, W., Pearce, N.: Trace element variations in coeval Holocene speleothems from GB Cave, southwest England. The Holocene, 9, 707–713, 1999.

Rimbu, N., Lohmann, G., Lorenz, S. J., Kim, J. H., & Schneider, R.R.: Holocene climate variability as derived from alkenone sea surface temperature and coupled ocean-atmosphere model experiments. Climate Dynamics, 23(2), 215-227, 2004.

Schimpf, D., Kilian, R., Kronz, A., Simon, K., Spötl, C., Worner, G., Deininger, M., Mangini, A.: The significance of chemical, isotopic, and detrital components in three coeval stalagmites from the superhumid southernmost Andes (53 degrees S) as high-resolution palaeo-climate proxies. Quat. Sci. Rev., 30, 443–459, 2011.

Shotyk, W., Weiss, D., Kramers, J. D., Frei, R., Cheburkin, A. K., Gloor, M., and Reese, S.: Geochemistry of the peat bog at Etang de la Gruère, Jura Mountains, Switzerland, and its record of atmospheric Pb and lithogenic trace metals (Sc, Ti, Y, Zr, and REE) since 12,370 14C yr BP, Geochim. Cosmochim. Acta, 65, 2337–2360, 2001.

Shotyk, W., Krachler M., Martinez-Cortizas, A., Cheburkin, A. K., and Emons, H.: A peat bog record of natural, pre-anthropogenic enrichments of trace elements in atmospheric aerosols since 12370 14C yr BP, and their variation with Holocene climate change, Earth Planet. Sci. Lett., 199, 21–37, 2002.

Shopov, Y.Y., Ford D.C., and Schwarcz, H.P.: Luminescent Microbanding in speleothems: High resolution chronology and paleoclimate. Geology, 22, 407-410, 1994.

Shindell, D.T., Schmidt, G.A., Miller, R.L. and Rind, D.: Northern Hemisphere winter climate response to greenhouse gas, ozone, solar, and volcanic forcing. *J. Geophys. Res*. 106, 7193–7210, 2001.

Smith, A.C., Wynn, P.M., Barker, P.A., Leng, M.J., Noble, S.R., and Tych, W.: North Atlantic forcing of moisture delivery to Europe throughout the Holocene. Scientific reports, 6, 2016.

Solanki, S.K., Usoskin, I.G, Kromber, B., Schüssler, M., Beer, J.: Unusual activity of the Sun during recent decades compared to the previous 11,000 years. Nature, 431, 1084–1087, 2004.

Soon, W., Velasco Herrera, V.M., Selvaraj, K., W., Traversi, R., Usoskin, I., *et al.:* A review of Holocene solar-linked climatic variation on centennial to millennial timescales: Physical processes, interpretative frameworks and a new multiple cross-wavelet transform algorithm. *Earth Sci. Rev.*, 134, 1–15, 2014.

Steinhilber, F., J.A. Abreu, J. Beer, I. Brunner, M. Christl, et al.: 9,400 years of cosmic radiation and solar activity from ice cores and tree rings. Proc. Natl. Academy of Sci. of U.S.A., 109, 16, 5967-5971, 2012.

Thompson, L. G., Mosley-Thompson, E., Davis, M. E., Henderson, K. A., Brecher, H. H., Zagorodnov, V. S., Mashiotta, T. A., Lin, P.-N., Mikhalenko, V. N., Hardy, D. R., and Beer, J.: Ice core records: evidence of Holocene climate change in tropical Africa, Science, 298, 589–593, 2000.

Tooth, A.F., Fairchild, I.J.: Soil and karst aquifer hydrological controls on the geochemical evolution of speleothem-forming drip waters, Crag Cave, southwest Ireland. J. Hydrol., 273, 51-68, 2003.

Treble, P., Shelley, J. M. G., and Chappell, J.: Comparison of high resolution sub-annual 781 records of trace elements in a modern (1911-1992) speleothem with instrumental climate data 782 from southwest Australia, Earth and Planetary Science Letters, 216, 141-153, 2003.

Treble, P. C., Fairchild, I. J., Baker, A., Meredith, K. M., Andersen, M. S., Salmon, S. U., Bradley, C., Wynn, P. M., Hankin, S., Wood, A., and McGuire, E.: Roles of bioproductivity, transpiration and fire in an eight-year record of cave dripwater chemistry from a forested catchment, southwest Australia. Geochim. Cosmochim. Ac., 184, 132–150, 2016.

Tremaine, D.M., and Froelich, P.N.: Speleothem trace elemental signatures: A hydrologic geochemical study of modern cave dripwaters and farmed calcite. Geochim. Cosmochim. Ac., 121, 522-545, 2013.

Tuner, T.E., Swindles, G.T., Charman, D.J., Langdon, P.G., Morris, P.J., Booth, R.K., Parry, L.E., Nicolas, J.E.: Solar cycles or random processes? Evaluating solar variability in Holocene climate records. Sci. Rep. 6, 23961; doi: 10.1038/srep23961 (2016).

Van Geel, B., Van Der Plicht, J., Kilian, M., Klaver, E., Kouwenberg, J., Renssen, H., Reynaud-Farrera, I., Waterbolk, H.: The sharp rise of Delta C-14 ca. 800 cal BC: Possible causes, related climatic teleconnections and the impact on human environments. Radiocarbon, 40, 535–550, 1996.

Verheyden, S., Keppens, E., Fairchild, I.J., Mc Dermott, F., Weis, D.: Sr isotope geochemistry of a Belgian Holocene speleothem: implications for paleoclimate reconstructions. Chemical Geology, 169, 131-144, 2000.

Verheyden, S.: Speleothems as palaeoclimatic archives. Isotopic and geochemical study of the cave environment and its Late Quaternary records, Unpubl, PhD Thesis, Vrije Universiteit Brussel, Belgium, 132 pp., 2001.

Verheyden, S., Baele, J.-M., Keppens, E., Genty, D., Cattani, O., Hai, C., Edwards, L., Hucai, Z., Van Strijdonck, M., and Quinif, Y.: The proserpine stalagmite (Han-sur-Lesse cave, Belgium): Preliminary environmental interpretation of the last 1000 years as recorded in a layered speleothem, Geol. Belg., 9, 245–256, 2006.

Verheyden, S., Genty, D., Deflandre, G., Quinif, Y., and Keppens, E.: Monitoring climatological, hydrological and geochemical parameters in the Pere Noel cave (Belgium): implication for the interpretation of speleothem isotopic and geochemical time-series, Int. J. Speleol., 37, 221–234, 2008.

Verheyden S., Keppens E., Van Strydonck M. & Quinif Y.: The 8.2 ka event: is it registered in Belgian speleothems? Speleogenesis, Online International Scientific Journal http://www.speleogenesis.info/journal/, issue 12, 2012.

Verheyden, S., Keppens, E., Quinif, Y., Cheng, H. J., and Edwards, L. R.: Late-glacial and Holocene climate reconstruction as inferred from a stalagmite – Grotte du Pere Noel, Han-sur-Lesse, Belgium, Geol. Belg., 17, 83–89, 2014.

Von Grafenstein, U., Erlenkeuser, H., Brauer, A., Jouzel, J., Johnsen, S.J.: A mid-European decadal isotope-climate record from 15,500 to 5000 years B.P. Science 284, 1654–1657, 1999.

White W.B.: Color of speleothems. In : Hill C.A. and Forti P. (Eds.): Cave minerals of the world. National Speleological Society: 239244, 1997.

Wang, Y., Cheng, H., Edwards, R. L., Kong, X., Shao, X., Chen, S., Wu, J., Jiang, X., Wang, X., and An, Z.: Millennial- and orbital-scale changes in the East Asian monsoon over the past 224,000 years, Nature, 451, 1090-1093, 2008.

Wagner, G., Beer, J., Masarik, J., Muscheler, R., Mende, W., Kubik, P. W., Laj, C., 5 Raisbeck, G. M., and Yiou, F.: Presence of the solar De Vries cycle ($\sim$ 205 years) during the last ice age, Geophys. Res. Lett., 28, 303–306, 2001.

Wanner, H., and Bütikofer, J.: Holocene Bond Cycles: real or imaginary? Geografie, 113, 338–350, 2008b.

Wanner, H., Beer, J., Butikofer, J., Crowley, T., Cubasch, U., Fluckiger, J., Goosse, H., Grosjean, M., Joos, F., Kaplan, J. O., Kuttel, M., Muller, S., Pentice, C., Solomina, O., Stocker, T., Tarasov, P., Wagner, M., and Widmann, M.: Mid to late Holocene climate change – an overview, Quaternary Sci. Rev., 27, 1791–1828, 2008.

Wanner, H., Solomina, O., Grosjean, M., Ritz, S. P., and Jetel, M.: Structure and origin of Holocene cold events, Quaternary Sci. Rev., 30, 3109–3123, 2011.

Wanner, H., Mercolli, L., Grosjean, M., and Ritz, S. P.: Holocene climate variability and change; a data-based review, J. Geol. Soc. London, 172, 254–263, 1, 2014.

Wynn, P.M., Fairchild, I.J., Baker, A., Frisia, S., Borsato, A., Baldini, J., McDermott, F.: Isotopic archives of sulphur in speleothems. Geochim. Cosmochim. Acta 72, 2465–2477, 2008.

Zhou, H.Y., Feng, Y.X., Zhao, J.X., et al.: Deglacial variations of Sr and $^{87}Sr/^{86}Sr$ ratio recorded by a stalagmite from Central China and their association with past climate and environment. Chem. Geol, 268, 233–247, 2009.

Table 1: Mass-spectrometric U/Th age data for the PN speleothem.

Distance from top of stalagmite (mm)	U(ppm)	^{234}U/^{238}U	^{230}Th/^{232}Th	^{230}Th/^{234}U	Age in years BP 1950	error +/-2σ	Laboratory
5	0.234	2.366	30	0.0185	2024	11	Open Univ-UK
45	0.297	2.391	59	0.0354	3907	16	Open Univ-UK
85	0.223	2.438	58	0.0518	5585	39	GET-FR
100	0.214	2.434	207	0.0501	5911	47	GET-FR
115	0.199	2.466	980	0.0585	6515	37	Open Univ-UK
170	0.181	2.462	335	0.0723	6461	48	GET-FR
190	0.168	2.497	1216	0.0613	6843	37	Open Univ-UK
250	2.781	2.496	2345	0.0641	7164	22	Open Univ-UK
307	0.199	2.492	1058	0.1066	7151	51	GET-FR
365	0.228	2.506	1902	0.0701	7853	45	Open Univ-UK
395	0.242	2.514	1401	0.0764	8232	39	Minnesota Univ-USA
425	0.238	2.552	1042	0.0810	9051	27	GET-FR
435	0.304	2.578	724	0.0816	9185	21	Open Univ-UK
490	0.269	2.595	1241	0.0888	9757	63	GET-FR
520	0.253	2.654	299	0.0912	10303	55	Open Univ-UK
635	0.179	2.729	161	0.1112	12671	71	Open Univ-UK
635	0.203	2.724	344	0.1117	12648	66	GET-FR

Decay constants used to calculate activity ratios from measured ratios are defined as follows: $\lambda^{238}U=1.551*10^{-10}$, $\lambda^{234}U=2.835*10^{-6}$, $\lambda^{230}Th=9.915*10^{-6}$, $\lambda^{232}Th=4.948*10^{-11}$. Age uncertainties are reported at the 2σ level.

Table 2 : Comparison between reference and measured values for five certified reference materials.

Standard	Element ($\mu g.g^{-1}$)	Ba	Sr	Mg	Al
NIST 610	Reference values	473 ±4	497,4 ±3,3	465±12,4	10800
	Mean measured values (n=7)	565 ±133	658 ±139	625±65	11458±215
	Reproducibility	76	79	90	98
NIST 612	reference values	37,74 ±1	76,15 ±0,5	77±4	11170
	Mean measured values (n=24)	45,1 ±9,7	82,6 ±6,6	78,9±8,3	10984±823
	Reproducibility	79	92	89	93
Nist 614	Reference values	3,15 ±0,05	45 ±0,4	34,39± 0,3	-
	Mean measured values (n=4)	3,5 ±0,8	43,4 ±1,6	38,1± 4	-
	Reproducibility	77	96	88	-
MACS-1	Reference values	114 ±8	219±20	10	110±16
	Mean measured values (n=15)	110 ±16	180±43	22	326±113
	Reproducibility	85	76	85	65
MACS-3	Reference values	58,7 ±2	6760 ±350	1756± 136	-
	Mean measured values (n=11)	47,2 ±12	6866 ±420	1566± 189	-
	Reproducibility	74	94	88	-
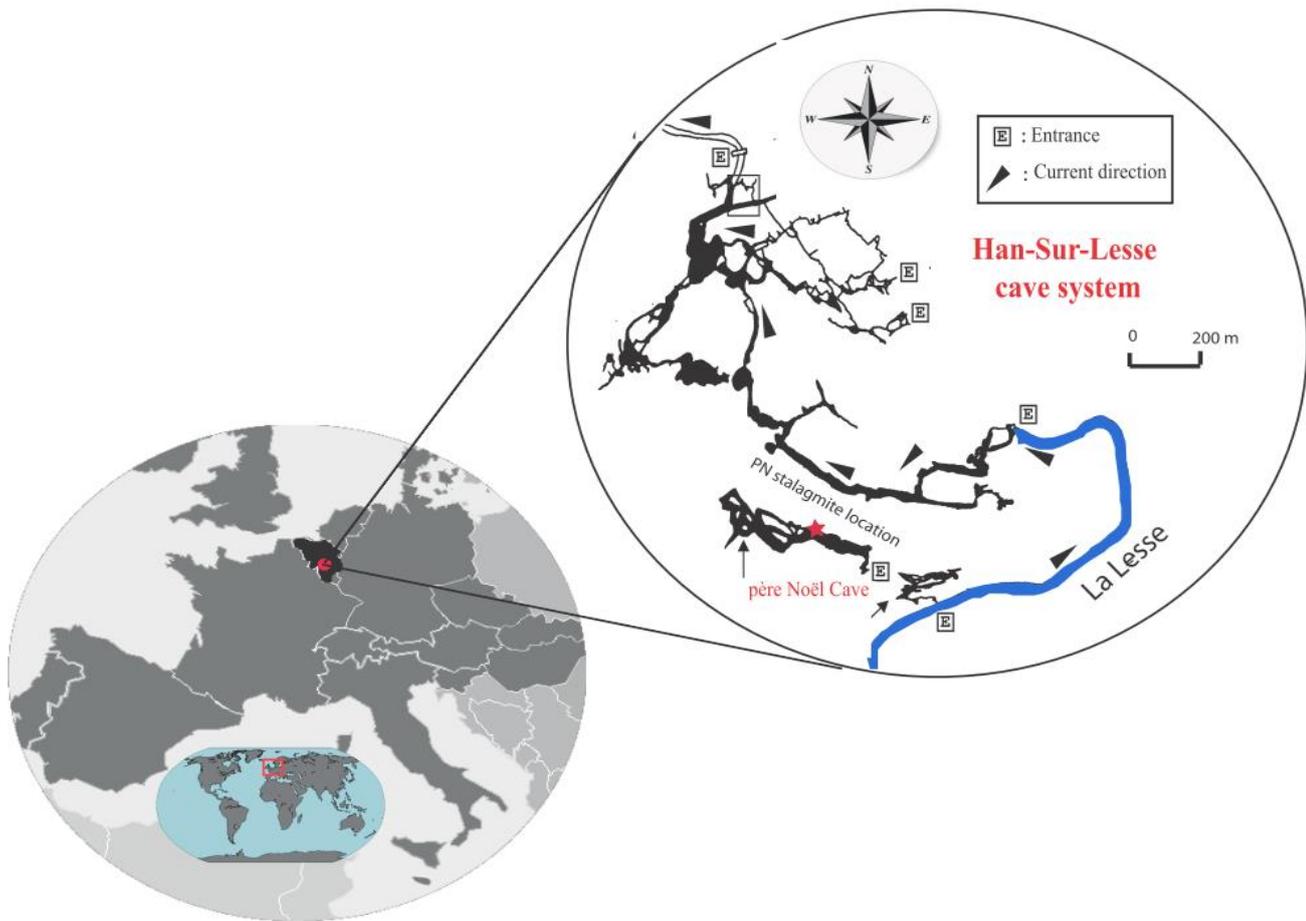
Figure 1: Location of the Père Noël Cave at Han-sur-Lesse, Belgium.
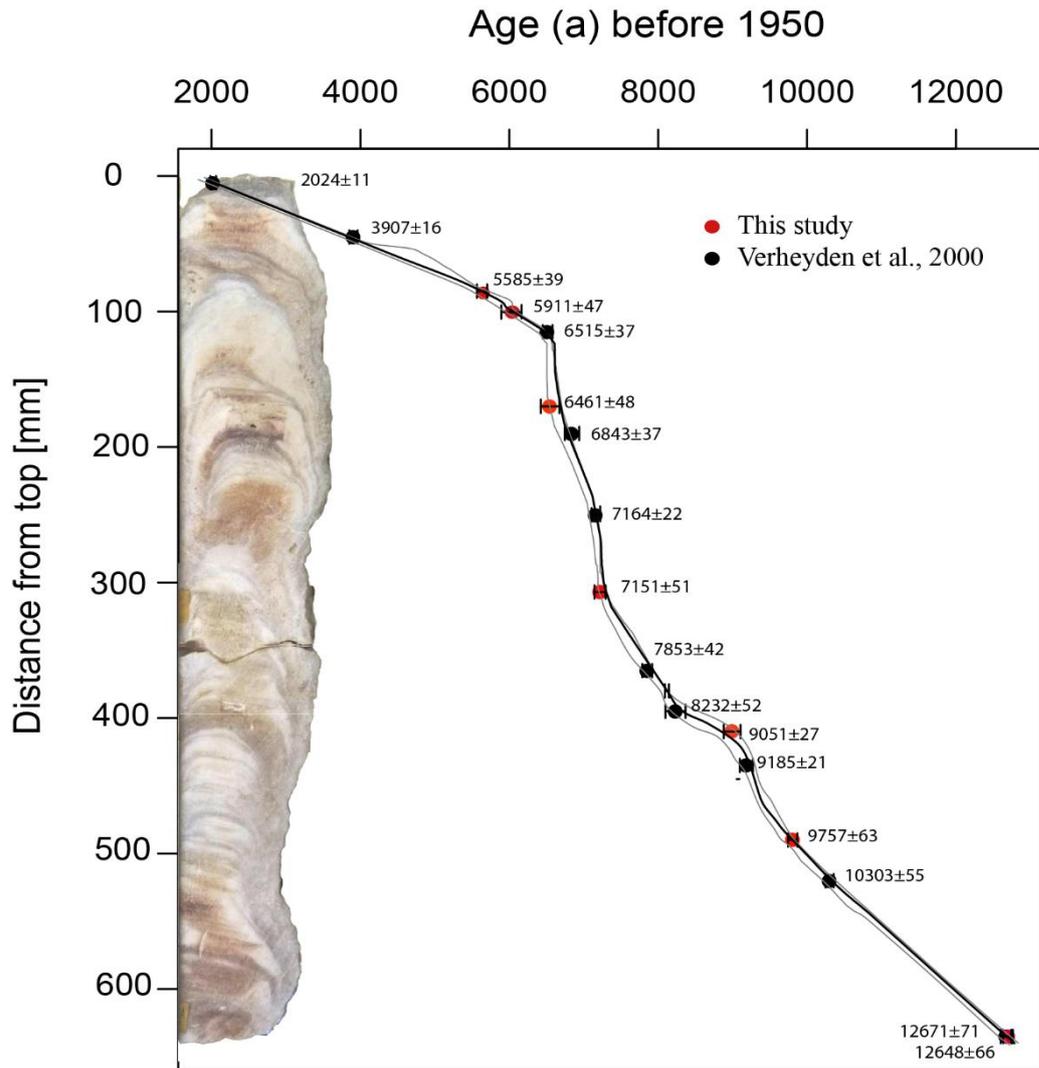
Figure 2: Age versus depth plots and average growth rate of the PN stalagmite. The black line represents the age model. Grey lines indicate the age dating uncertainties. Red and black dots present the U/Th dates.
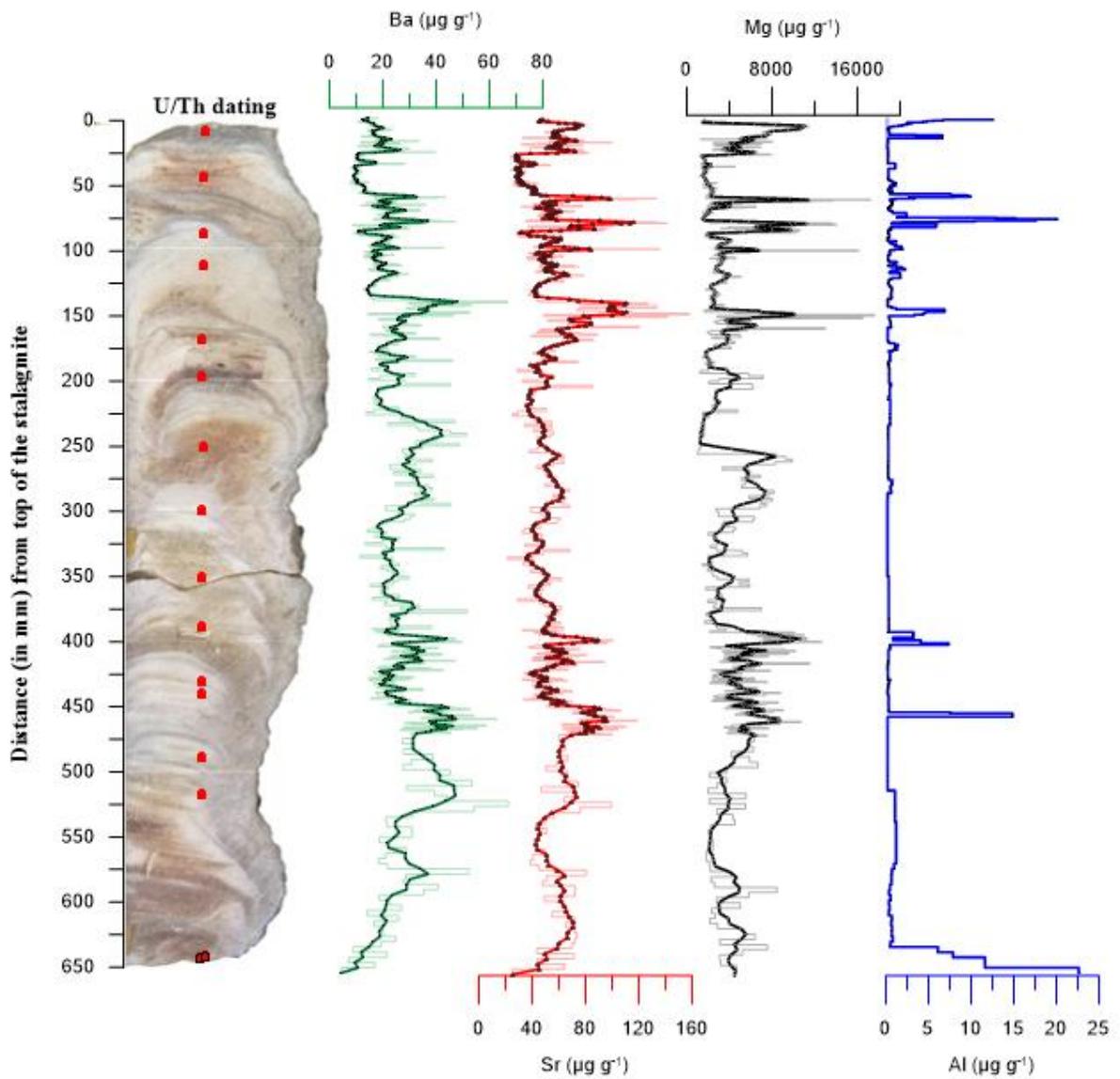
Figure 3: Stalagmite PN laser ablation Sr (red line), Ba (green line), Mg (black line) and Al (blue line) concentrations record for 64 cm-length. The measurement was made with 300-1000 µm intervals (light color). Dark line color presents mean of three measurements.
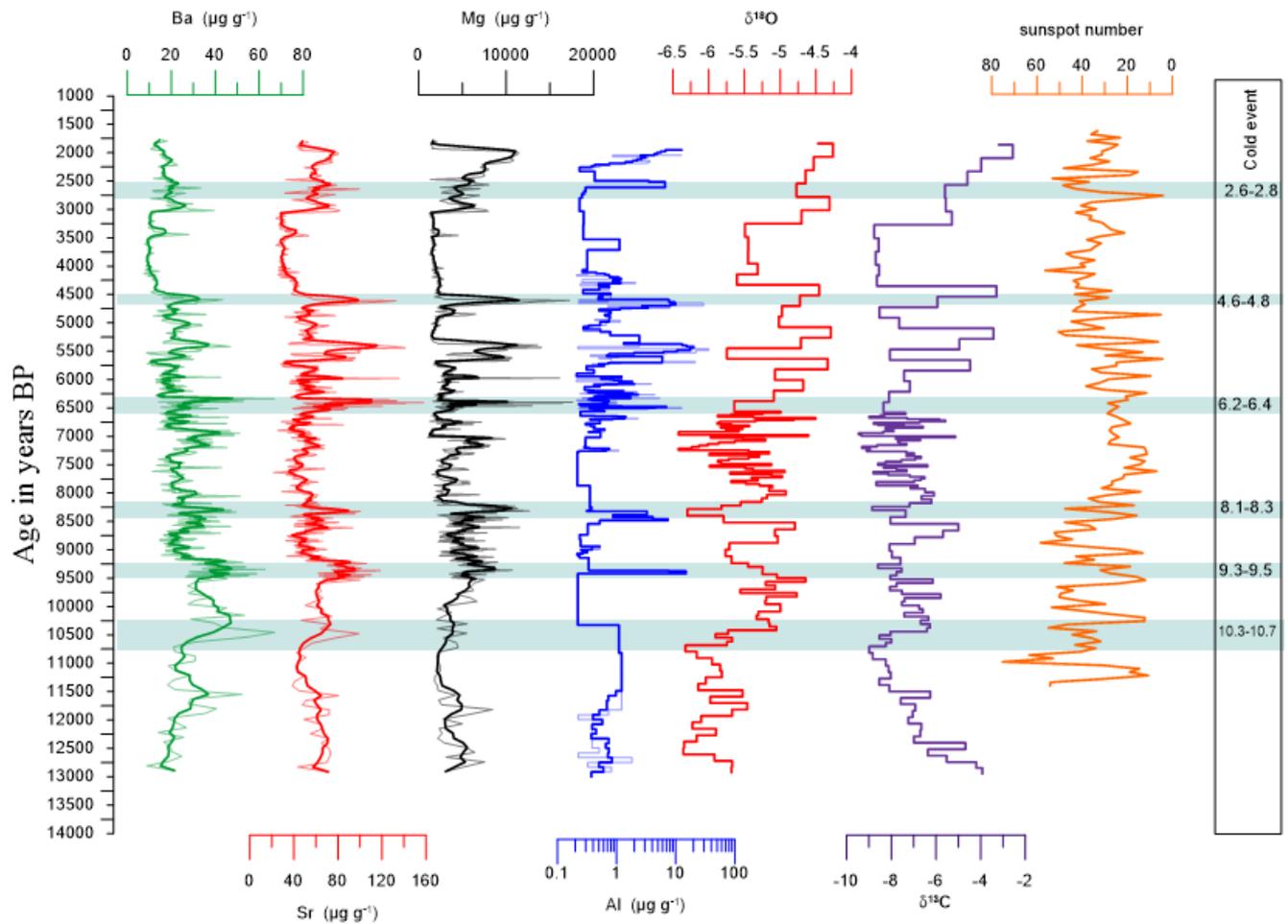
Figure 4: Stalagmite PN laser ablation Sr (red line), Ba (green line), Mg (black line) and Al (blue line) concentrations records for the period 12.7 to 1.8 ka BP. The dark color lines presents mean of three measurements. PN Stalagmite $\delta^{18}O$ (red line) and $\delta^{13}C$ (purple line) measurements were made every 5 mm. Orange line shows sunspot number related to solar activity (Solanki et al., 2004).
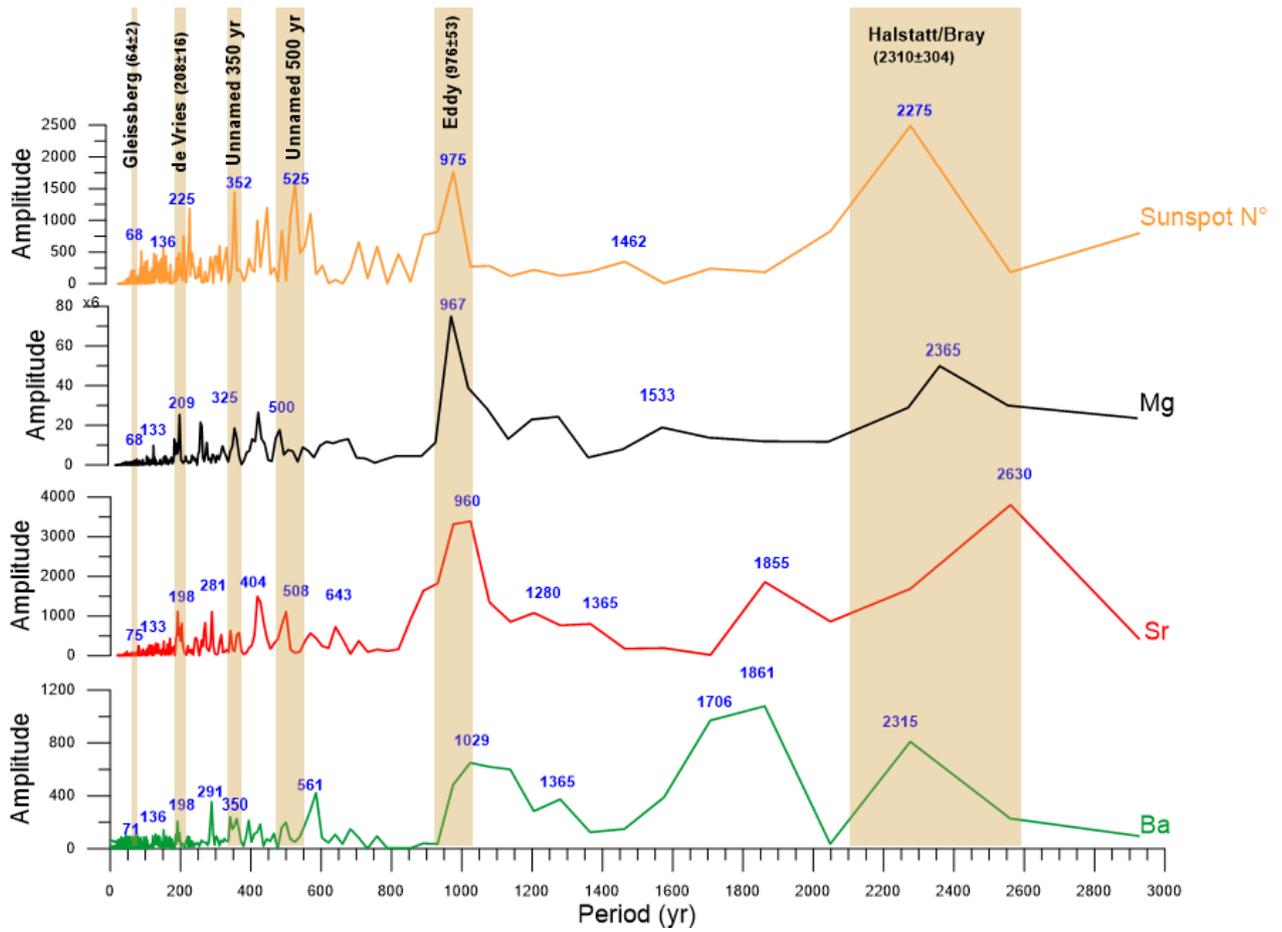
Figure 5: Intensity of cycles in PN trace elements and sunspot activity (Solanki et al., 2004). The power spectra demonstrate that the strongest cycles of PN trace elements are with durations of 68-75, 133-136, 198-209, 291-358, 404-602,912-1029 and 2365-2670 yr.
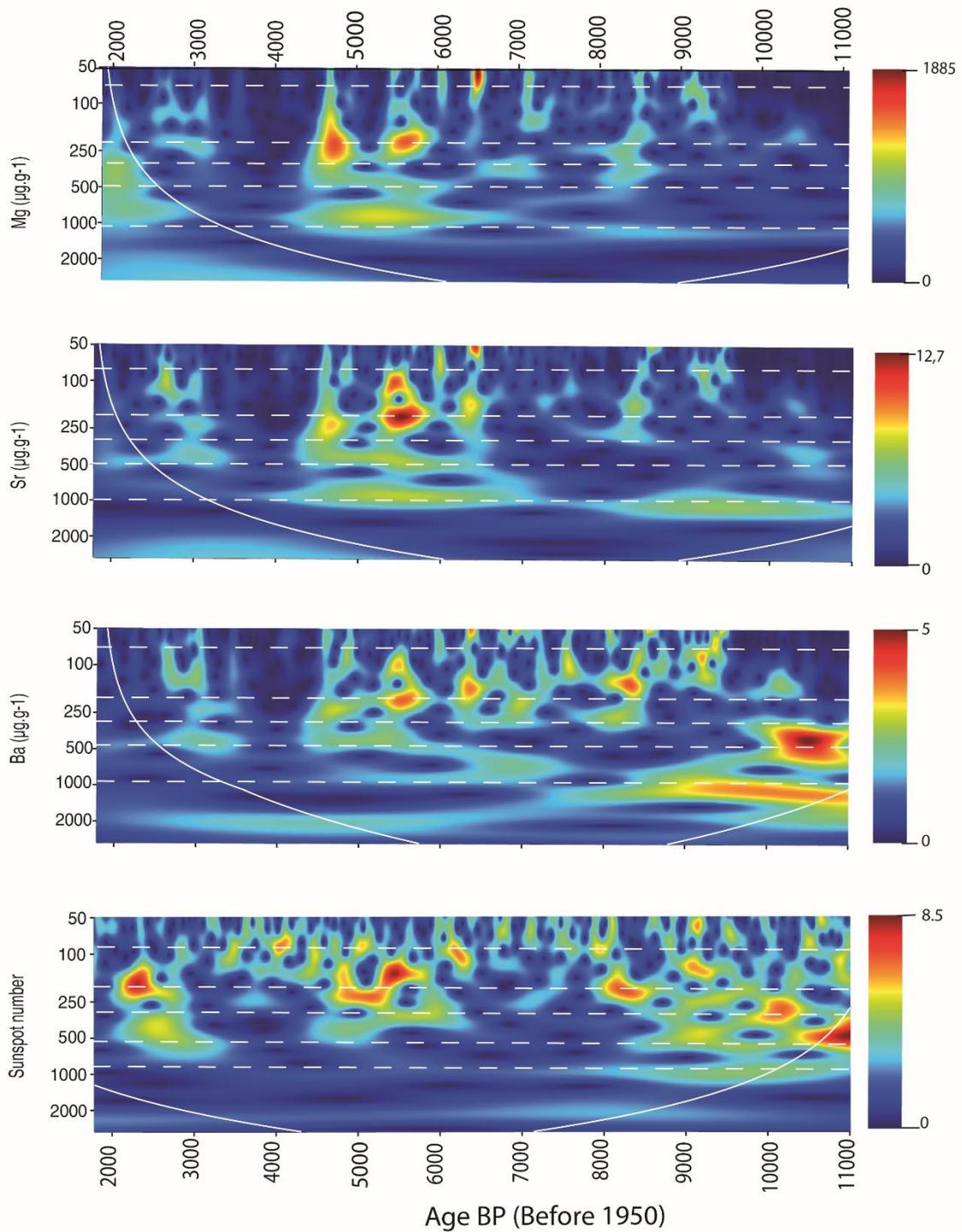
Figure 6: Continuous wavelet transform spectra for sunspot number (Solanki et al., 2004), Ba, Sr, Mg. Spectral power (variance) is shown by colors ranging from deep blue (weak) to deep red (strong). Ba, Sr, and Mg concentrations and sunspot number are presented along y-axis. The white contour lines represent 95% confidence level.
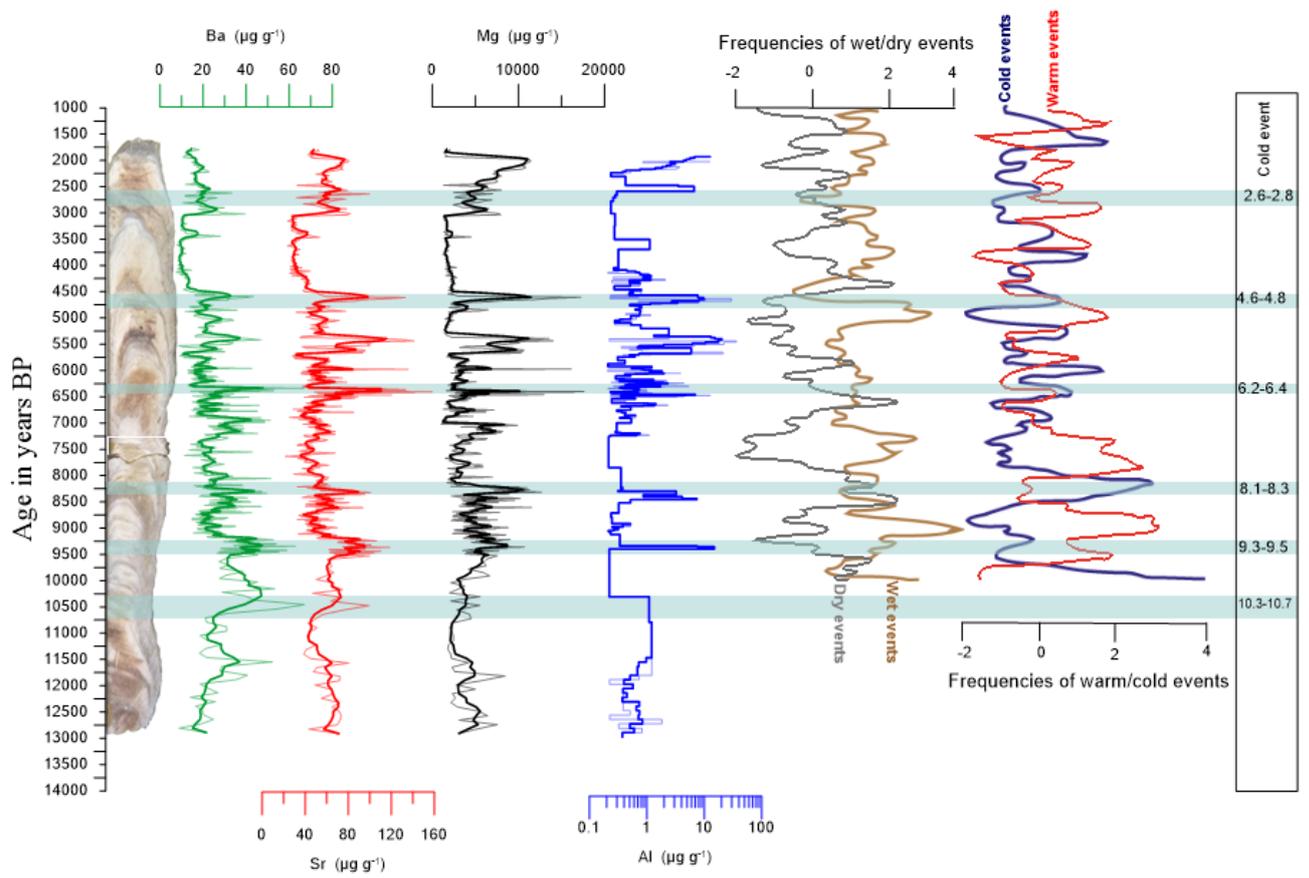
Figure 7: A comparison between PN trace elements contents (Sr = red line; Ba = green line; Mg = black line; and Al = blue line), frequency of warm/cold and wet/dry events in the Northern Hemisphere as defined by Wanner et al. (2014). The blue horizontal bars mark cold events.

Supplementary data

Continuous wavelet transform

In this section we describe step-by-step our wavelet-induced mode extraction. First, since the raw trace element records consist in unevenly spaced time series, each dataset is re-sampled at the frequency of one observation every ten years and centered so that it has zero mean. Prior to perform the spectral analysis each dataset was resampled as follows : for each age t (in BP) in the evenly spaced sequence 1789, 1799, 1809,..., the corresponding concentration of trace element is obtained through a linear interpolation between the two consecutive data points whose ages (in BP) surround t. Since 92% of the time gaps in the original time series are lower than 25 years and the remaining 8% all occur at once at the end of the signal (after 9.5 ka BP), the effects of such a resampling on the results presented are negligible (especially for periods such as the de Vries cycle or larger)(Fig.6, 7, 8). The periodograms of the resampled time series displayed in Fig.5 (see text) are computed using the periodogram function in Matlab.

The rest of this work is based on the continuous wavelet transform (CWT); self-made codes implemented for Scilab are available upon request (or can be derived from the WIME toolbox). The main wavelet-related principles involved in this study are inspired from (Deliège and Nicolay, 2017; Deliège et al., 2016; Nicolay S., 2011; Nicolay et al., 2009) and are summarized below. More information about wavelets and the CWT can be found in e.g. (Daubechies I., 1992; Mallat S., 1999; Meyer Y., 1993; Torrence and Compo, 1998).

Given a wavelet ψ and a function f, the wavelet transform of f at time t and at scale $a > 0$ is defined as:

$$W_f(t,a) = \int f(x)\bar{\psi}\left(\frac{x-t}{a}\right)\frac{dx}{a}$$

where ψ^- is the complex conjugate of ψ. In the context of time-frequency analysis, it is recommended to use a wavelet well-located in the frequency domain (Nicolay S., 2011). In this work, we use the wavelet ψ defined by its Fourier transform as:

$$\hat{\psi}(\nu) = \sin\left(\frac{\pi\nu}{2\Omega}\right)e^{\frac{-(\nu-\Omega)^2}{2}}$$

with $\Omega = \pi\sqrt{2}/\ln 2$, which is similar to the Morlet wavelet but with exactly one vanishing moment (Torrence and Compo, 1998).

Since $\left|\hat{\psi}(\nu)\right| < 10^{-5}$ if $\nu \leq 0$, ψ can be considered as a progressive wavelet (i.e. is zero for negative arguments) and thus allows an easy recovery of trigonometric functions. Indeed, if $f(x) = \cos(\omega x)$, then

$$W_f(t,a) = \frac{1}{2}e^{it\omega}\overline{\hat{\psi}(a\omega)}$$

Therefore, for a fixed time t, if a^* stands for the scale at which the function $a \rightarrow |W_f(t,a)|$ reaches its maximum, then we have $a^*\omega = \Omega$. Consequently, the value of ω can be obtained (if unknown) and f is recovered with the real part of its CWT: $f(x) = 2\Re(W_f(x,a^*))$. Drawing on this principle, the general scheme for extracting the dominant components of a given function f is the following:

1.   The CWT of f is computed and the application $(t,a) \rightarrow |W_f(t,a)|$ can be seen as a time-frequency representation of f.

2.   The wavelet-based spectral composition of f, also called wavelet spectrum of f, i.e. the wavelet counterpart of its Fourier spectrum, is obtained as

$\Lambda : a \to E_t |W_f(t,a)|$, where E_t denotes the mean over time.

3.   For each scale s at which Λ reaches a local maximum, a component c_s is extracted from f as $c_s(x) = 2\Re(W_f(x,s))$.

By adding the modes obtained this way, the CWT allows an almost complete reconstruction of f with smooth amplitude modulated-frequency modulated (AM-FM) components. However, it is slightly different in practice because the signal considered has a finite length and thus has to be padded at its edges. The chosen padding (zero-padding in the present case) irremediably flaws a proportion of the wavelet coefficients, which creates a so-called cone of influence (see e.g. Deliège and Nicolay, 2016). Even though it is generally impossible to correct exactly the border effects, noticeable improvements can be made to sharpen the edges of the components extracted and the reconstruction of the initial signal by iterating the process described above. More precisely, the technique used in this work in complement to the previous algorithm is the following.

Let us imagine that the components of interest are located at scales $(s_i)_{i \in I}$ (for some set of indices I). Then, at the first iteration of steps 1 to 3 we extract $(c^1_i)_{i \in I}$ as $c^1_i(x) = 2\Re(W_f(x,s_i))$. We now perform the CWT and the extraction at the same scales $(s_i)_{i \in I}$ but with

$$f_1 = f - \sum_{i \in I} c^1_i$$

and get the modes $(c^2_i)_{i \in I}$, then again repeat the process with $f_2 = f_1 - \sum_{i \in I} c^2_i$, and so on. The iterations stop when the components extracted are not significant anymore, i.e. at iteration J if

$$\max\left\{\left\|c^J_i\right\|, i \in I\right\} < \alpha \ \max\left\{\left\|c^1_i\right\|, i \in I\right\}$$

where $\|.\|$ denotes the energy (square of L^2 norm) of a signal and α is a threshold chosen as 0.01. The final components $(c_i)_{i \in I}$ are then obtained as:

$$c_i = \sum_{j=1}^{J} c^j_i.$$

Although most of the energy of f has already been drained at the end of the procedure described in this section, it may turn out useful to examine what kind of information still remains in f. For that purpose, the whole method can be applied to $f - \sum_{i \in I} c_i$ to find out which frequencies might have been hidden behind the dominant ones and thus obtain "secondary components", if any.

References

Daubechies I.: Ten Lectures on Wavelets. SIAM, 1992

Deliège, A. and Nicolay, S.: Extracting oscillating components from nonstationary time series: A wavelet-induced method, to appear (accepted) in Physical Review E, 2017. DOI:10.1103/PhysRevE.00.003300.

Deliège, A. and Nicolay, S.: A New Wavelet-Based Mode Decomposition for Oscillating Signals and Comparison with the Empirical Mode Decomposition, 13th International Conference on Information Technology, 959-968, 2016.

Mallat, S.: A Wavelet Tour of Signal Processing. Academic Press, 1999.

Meyer, Y.: Wavelets and Operators, volume 1. Cambridge University Press, Cambridge, 1993.

Nicolay, S.: A wavelet-based mode decomposition. European Physical Journal B, 80, 223-232, 2011.

Nicolay, S., Mabille, G., Fettweis, X., and Erpicum, M.: 30 and 43 months period cycles found in air temperature time series using the Morlet wavelet method. Climate dynamics, 33(7),1117-1129, 2009.

Torrence, C. and Compo, G.: A practical guide to wavelet analysis. Bulletin of the American Meteorological Society, 79, 61-78, 1998.

Original data (PN trace element contents, sunspot number, and temperature) were filtered according to the dominant periods revealed by the CWT (Fig.1-5).
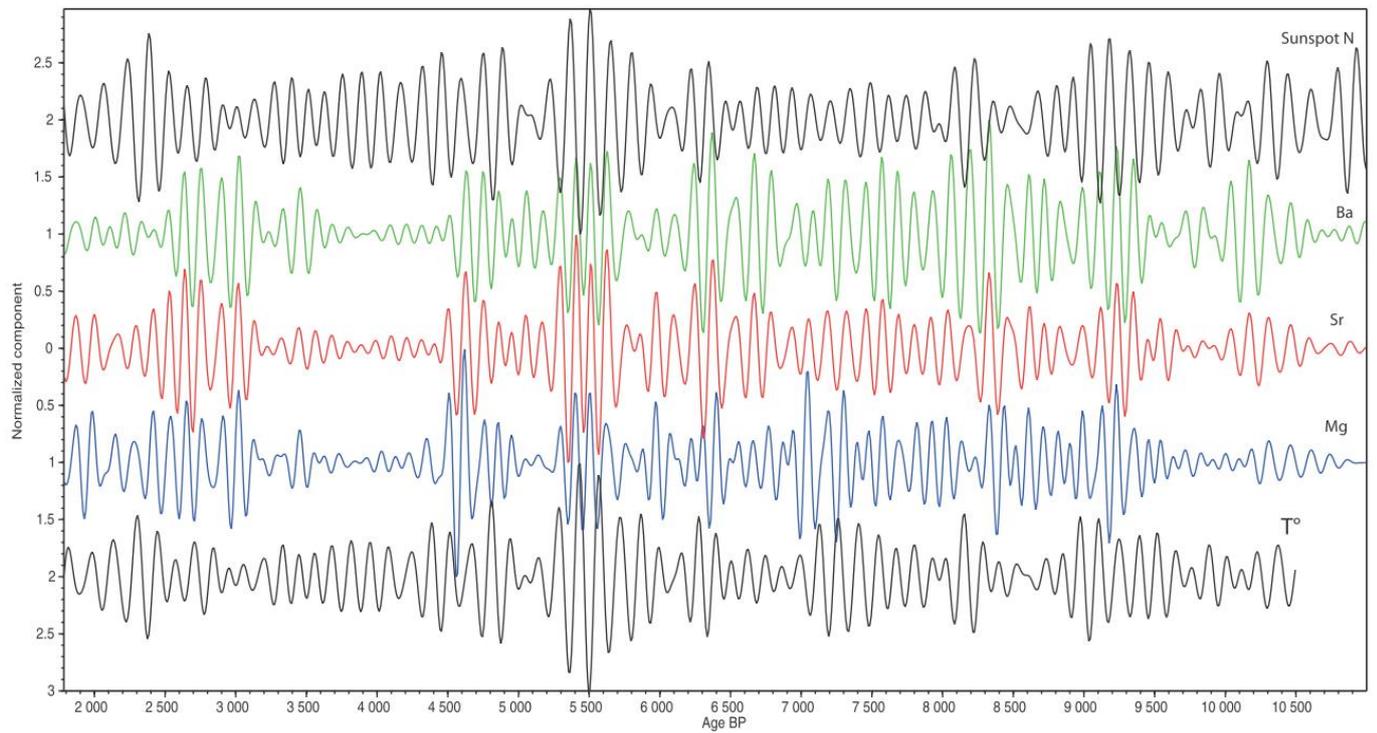
**Figure 1: the Gleissberg cycle (70-100 yr)**
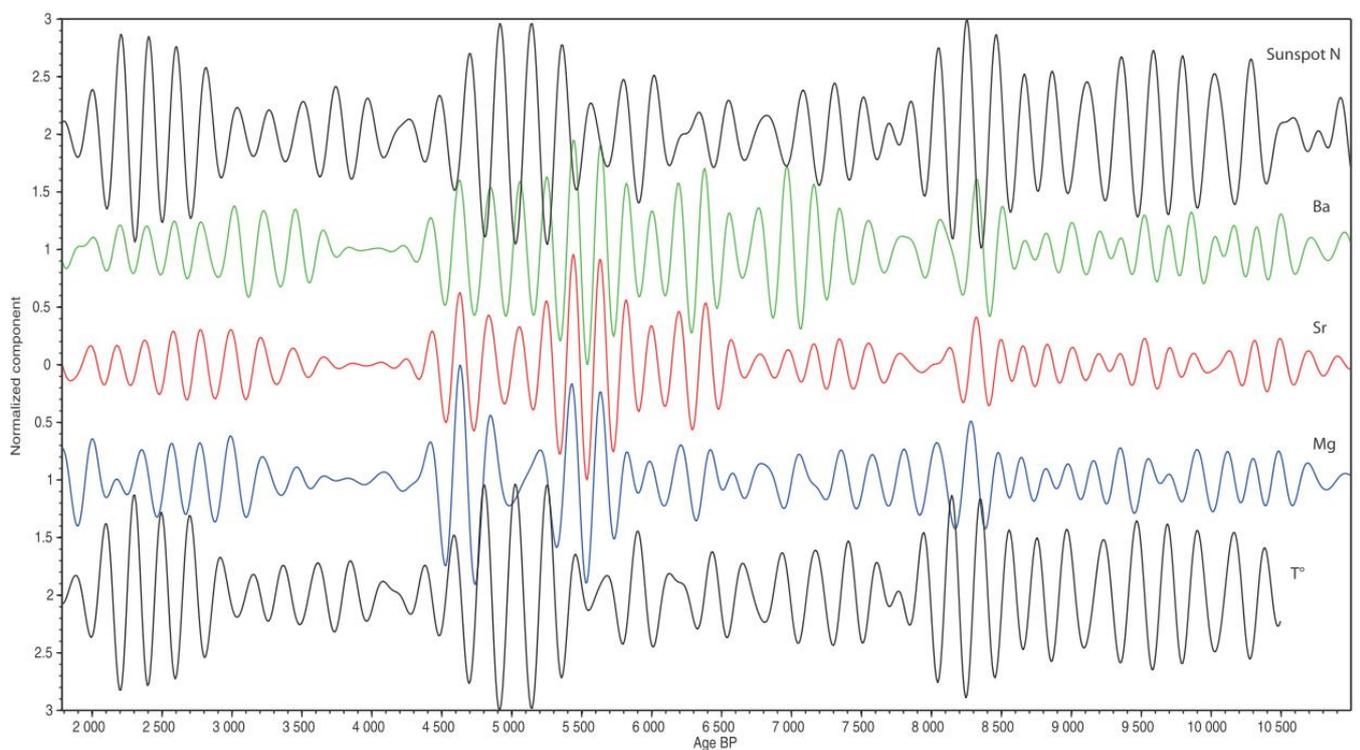

Figure 2: de Vries cycle (200-210 yr)
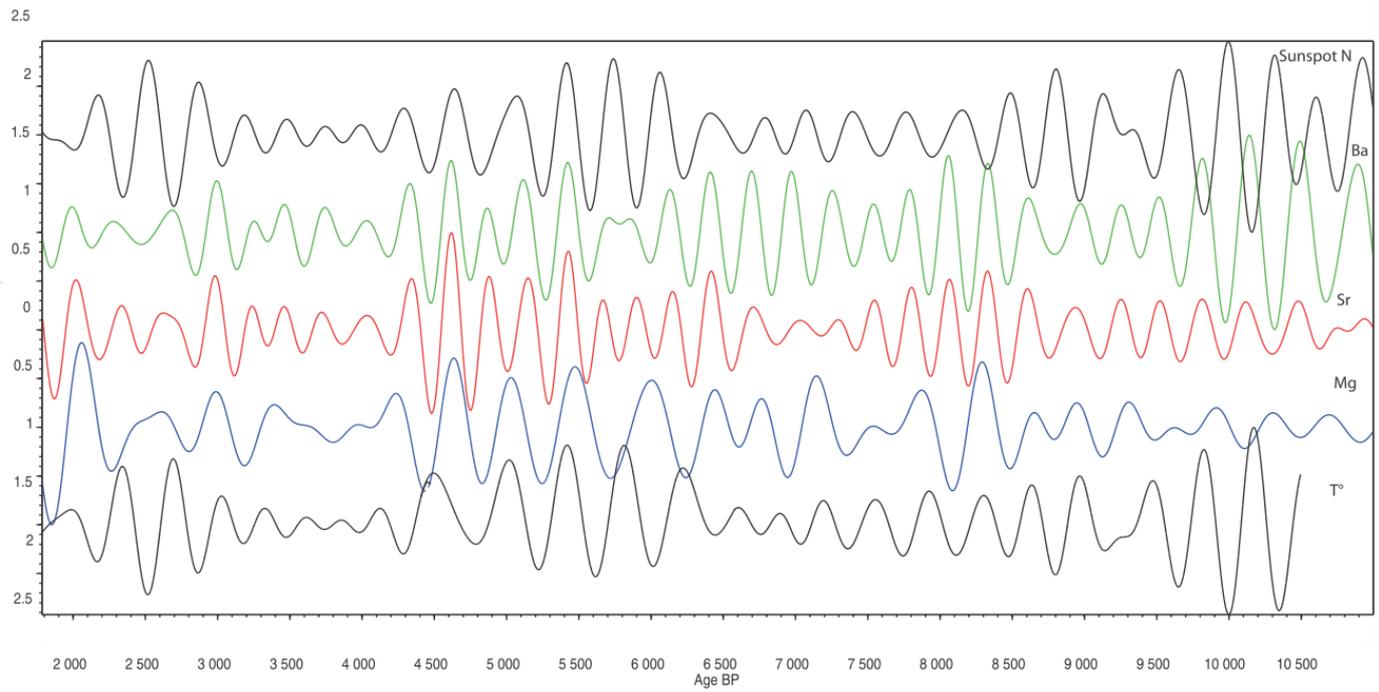
**Figure 3: Unnamed cycles 350 yrs**
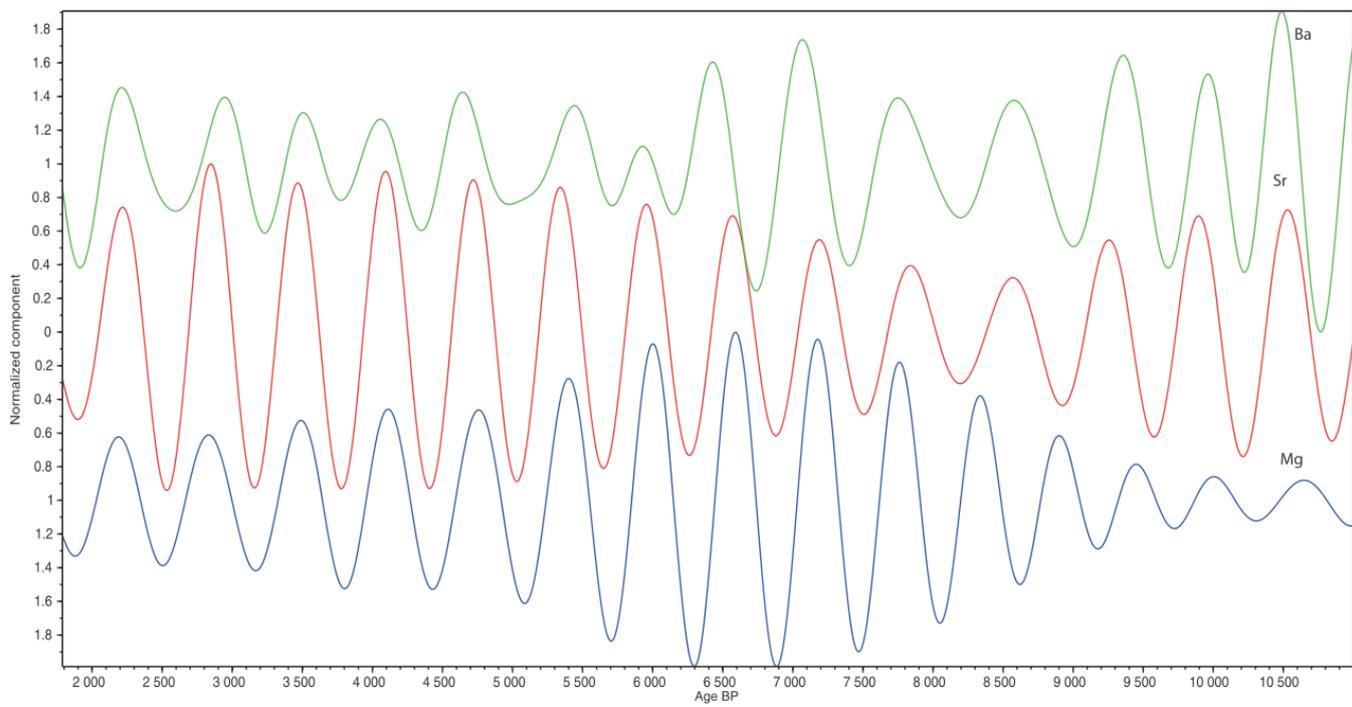
Figure 4: Unnamed cycles 550 yrs
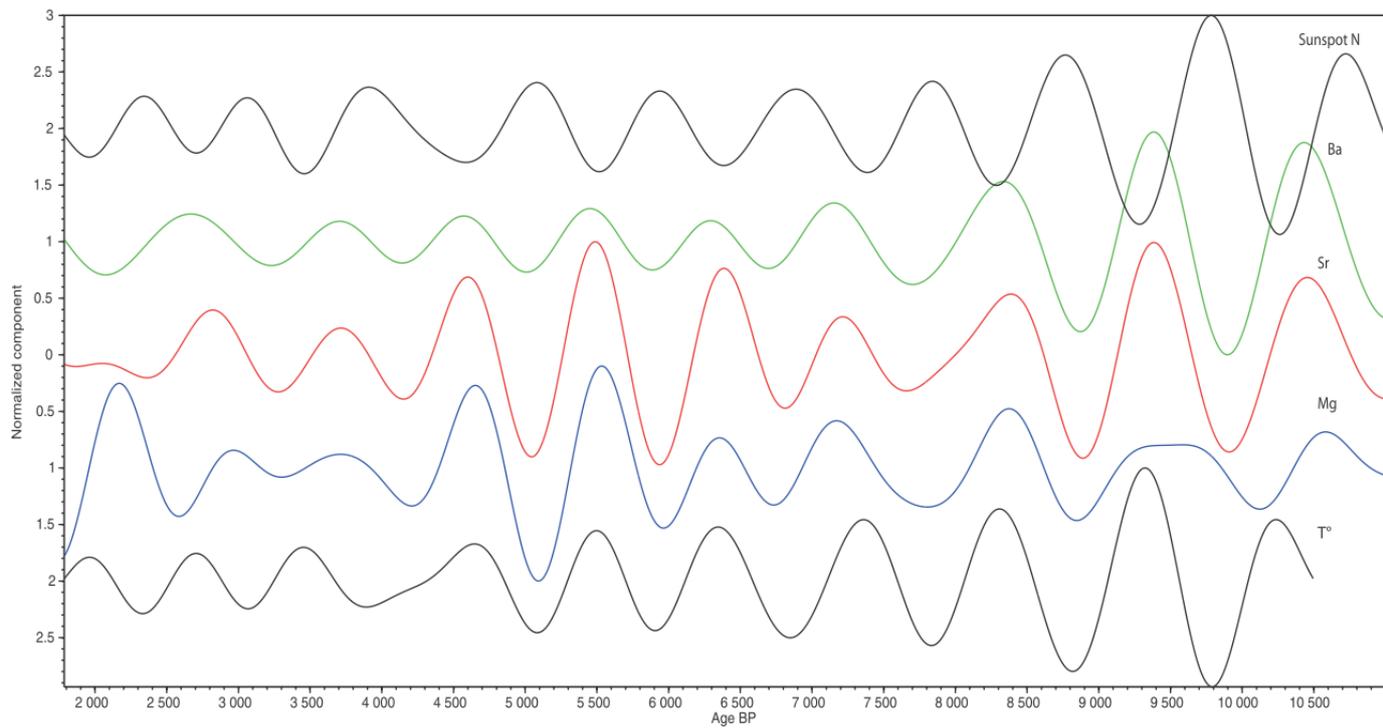
Figure 5: Eddy cycle (1000 yr)
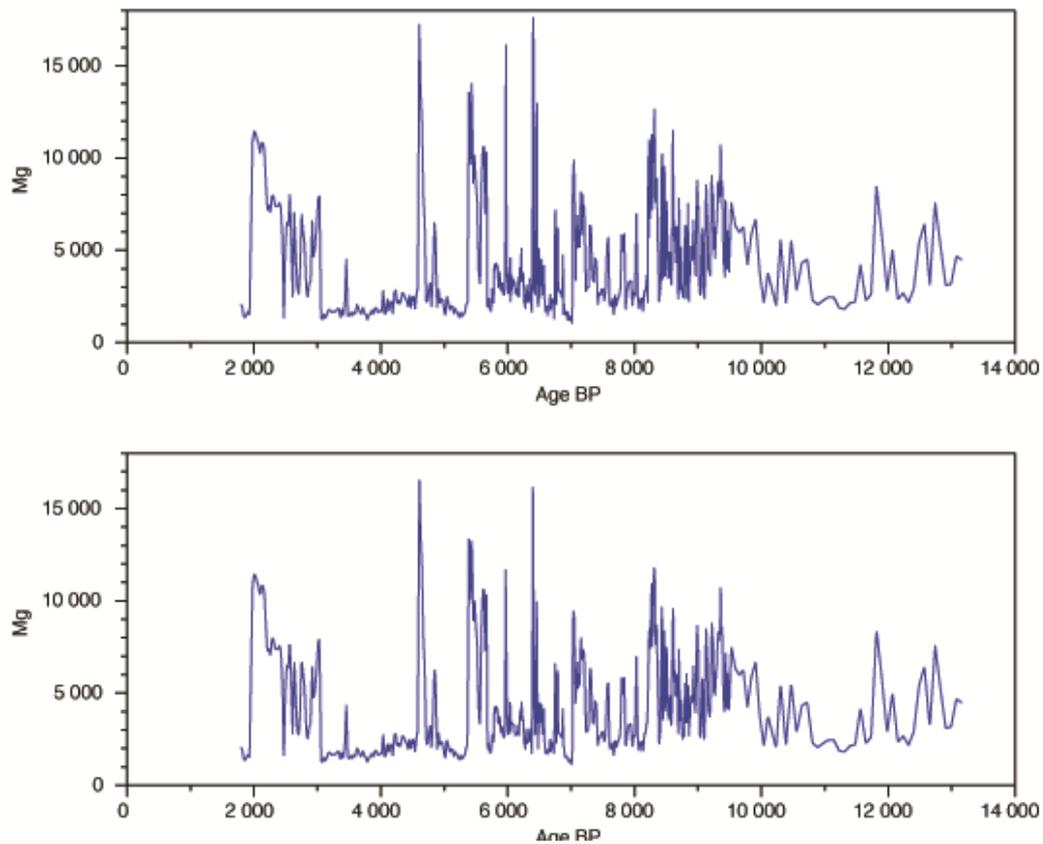
Fig 6: Top: initial signal for the trace element Mg (unevenly spaced time series). Bottom: resampled signal (evenly spaced time series, one data point every ten years).
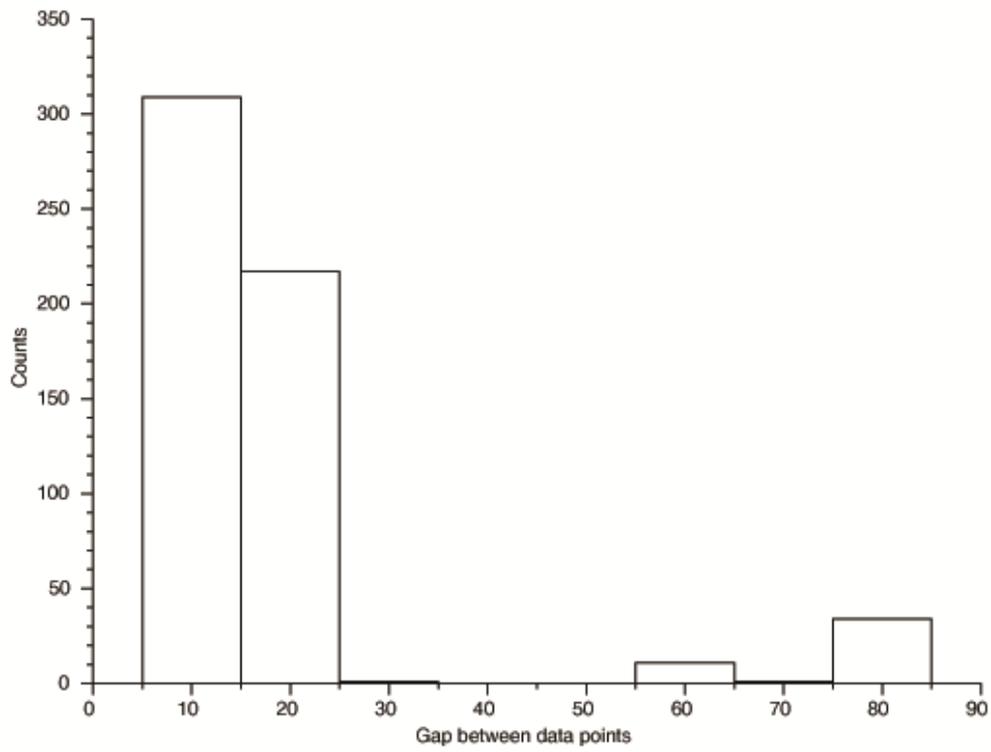
**Fig 7: Histogram of the time gaps between consecutive data.**
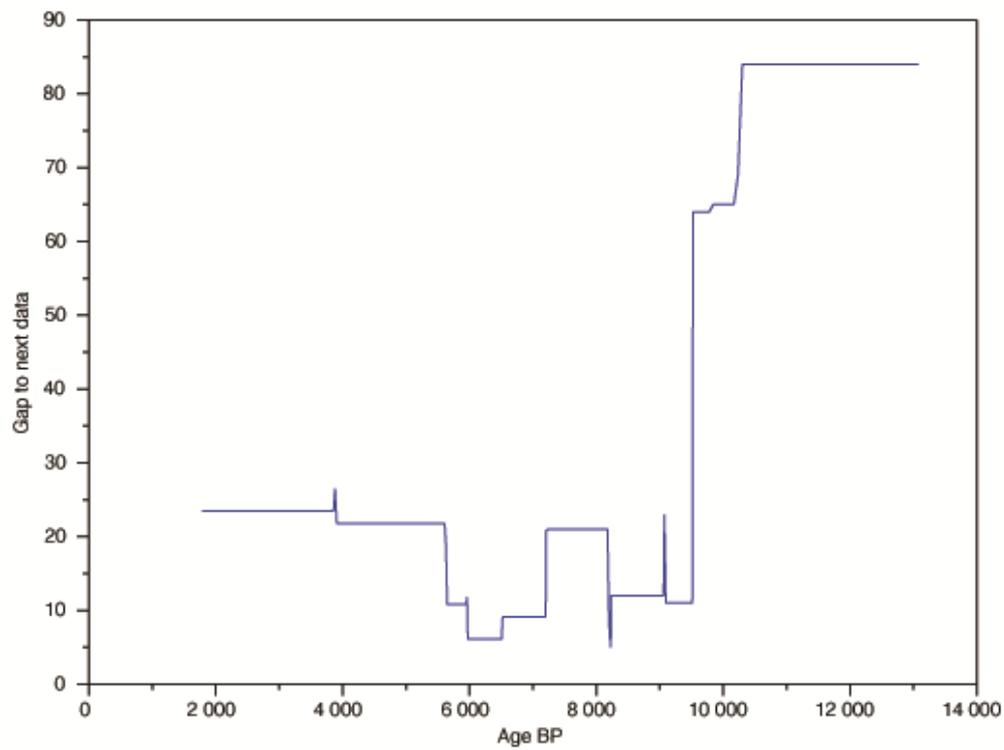
Fig 8: Time gap between consecutive data points as function of the age BP. For example, the first and second ages measured are 1789 and 1812.5, thus the first point of the curve is (1789, 23.5).