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The age of ambergris

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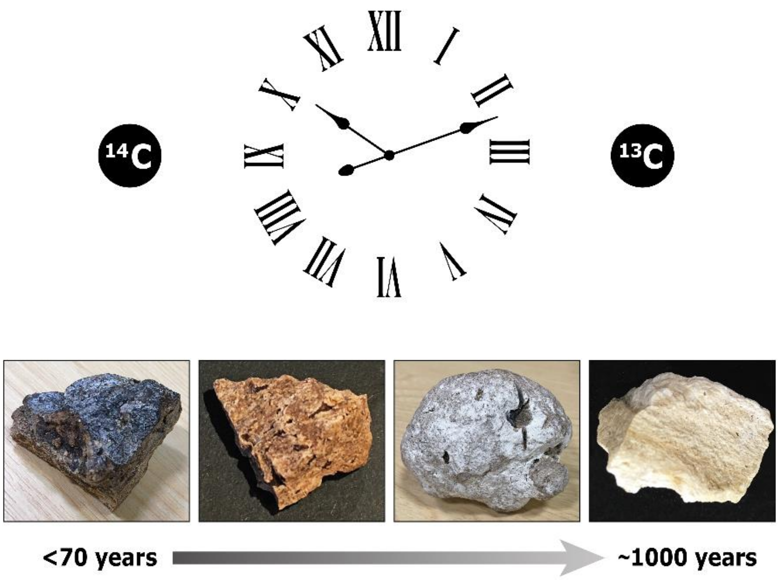

Ambergris, which is a coprolith originating from the sperm whale, has been found only rarely, but for centuries, as jetsam on beaches all over the world. There are no reliable data indicating how long such samples may have remained at sea, with unsubstantiated accounts suggesting maybe decades. Here, we obtained over forty jetsam samples many collected on known dates, from mostly known beach locations across the globe. Such an inventory of verified jetsam ambergris is unprecedented. Each sample was characterised by analytical methods such as gas chromatography-mass spectrometry (GC-MS). We then determined the radiocarbon ages of some of the samples by well-described accelerator-MS techniques. Surprisingly, some samples of jetsam have remained in the environment for about a thousand years.

1. Introduction

Ambergris is an enigmatic waxy substance, known since the ninth century (Levey 1961; Clarke 2006; Read 2013; Srinivasan 2015; Brito et al. 2016; Azzolini 2017). It was once a global economic commodity (Brito et al. 2016) and, for example, was long considered valuable as incense. In perfumery it was used in pomanders and to perfume gloves, including for Queen Elizabeth I and Catherine de Medici (Dugan 2011).

Ambergris has been known to occur in the sperm whale (*Physeter macrocephalus*), since at least the time of Marco Polo in the 13th Century, and was certainly known at the time of Elizabeth I (Purchas 1613; Bolyston 1724). Indeed, it has been used to anoint English monarchs from the time of King Charles I in 1626, to the present Queen Elizabeth II in 1953.

For a long time, its origin was a matter of debate. A series of questions raised by the Royal Society of London during the mid- to late-seventeenth century included two relating to the origins of ambergris (Read 2013; Dugan 2011). Finally in 1783, Dr Franz-Xavier Schwediawer and Sir Joseph Banks determined that ambergris is a natural product of the sperm whale and not simply ingested by the animal (Schwediawer and Banks 1783): a view which has ever since, been accepted (Clarke 2006).

In more modern times, data from whale catches (Berzin 1971; Korzh and Strigina 1972; Clarke 2006) supported much earlier reports (Boylston 1724) that ambergris only actually occurs in about one in a hundred whales. To date, sperm (and probably pygmy and dwarf sperm) whales (Clarke 2006), are the only known natural sources. Although historically it was important (Brito et al. 2016), ambergris is now largely a rare biological and chemical curiosity; synthetic chemical analogues have mostly replaced the natural material in perfumery (Serra 2013). Unsurprisingly therefore, few modern investigations of the composition and fate of this mammalian coprolith have been made.

However, it is known that ambergris taken historically from whales and now archived in museums, contains the terpenoid alcohol, ambrein and significant, though variable, proportions of a number of faecal steroids (Baynes-Cope 1962; Rowland and Sutton 2017).

For centuries, ambergris has also been found as jetsam on beaches from all over the globe (Boylston 1724; Clarke, 2006). Although verified accounts are virtually absent from the scientific literature, with unsubstantiated media accounts predominating (Kemp 2012), recent sparse data have shown that, in contrast to whale ambergris, extracts of jetsam samples comprise almost entirely ambrein, with, so far, very low steroid contents (Rowland and Sutton 2017; Rowland et al. 2018). However, very few data exist and there are no reliable data indicating how long such jetsam ambergris samples may have remained in the environment, with unsubstantiated accounts suggesting years, or maybe decades (Kemp 2018).

Here we obtained over forty rare jetsam ambergris samples collected by ourselves, and by others, many on known dates and from mostly known beach locations across the globe. Each sample was characterised by determining its chemical composition by analytical methods such as gas chromatography-mass spectrometry (GC-MS) using the methods of Rowland and Sutton (2017) and of Rowland et al. (2018). We also determined the radiocarbon (^{14}C) ages of the samples by well-described accelerator-MS (AMS) techniques (e.g. Synal et al. 2007; Wacker et al. 2010). AMS also provided approximate measures of the $^{13}C/^{12}C$ isotope ratios of the ambergris.

2. Results and discussion

We verified that each extract of jetsam (Table 1) contained ambrein as a major constituent of the organic soluble portions, by previously published methods of GC-MS and Fourier transform infrared spectroscopy (Rowland and Sutton 2017).

Samples included (Figure 1) those from the northern hemisphere (e.g. Japan, England, Scotland, Ireland, France, The Bahamas), nearer the equator (Somalia, Kenya, Sri Lanka, Indonesia) and from the southern hemisphere (Chile, New Zealand, Australia). Some pieces of ambergris reportedly weighed over twenty kilograms (Table 1). The samples were found to comprise mainly material extractable into dichloromethane (mean 96 ± 7 %; n=43); of this, most GC-MS detectable material was ambrein (mean 81 ± 22 %; n=43).

Accelerator mass spectrometry (AMS) analyses showed that the $^{13}C/^{12}C$ isotope ratios in the ambergris ranged from -15 to -25 ‰ (Table 1; mean -21 ± 2 ‰; n=26). Repeat analyses of samples 6-9 from Chile (Table 1; n=4) established the reproducibility of the method for replicate samples as ± 0.4 ‰.

The radiocarbon dates of the jetsam ambergris were also determined by AMS. The data indicated that four jetsam ambergris samples collected from Mar Brava beach, Chiloé Island, Chile in 2017, had a radiocarbon age of 1538 ± 14 y (Table 1). A further sample had a radiocarbon age of 1500 y (Table S1 and Figure S2). Calibration of radiocarbon age requires knowledge of any ‘reservoir effects’ of ^{14}C in the system under study. Since such ‘reservoir effects’ for sperm whale carbon are presently unknown, we calibrated the radiocarbon age using the Marine13 calibration curve (Reimer et al. 2013). This revealed that the samples from Chile were about 1000 years old (Table 1; Figure S1). Two of the other samples (samples 10 and 11; Table 1) had radiocarbon dates of 545 years and 219 years. Use of the same calibration (Figure S1) indicated an age for sample 10 of between 184 and 328 years. The ^{14}C data for the other samples (Table 1) indicated that they were ‘modern’ ($F^{14}C$ values up to 1.057). ‘Modern’ samples are those which demonstrate the incorporation of ^{14}C resulting from atmospheric nuclear weapons testing which started in the 1950s and thus indicate a modern post-1950s origin for the carbon. Indeed, as verification, an ambergris sample taken from a dead sperm whale beached in The Netherlands (Rowland and Sutton 2017) in 2012, showed a $F^{14}C$ value of 1.062 (Table S1).

The major organic constituent of ambergris, ambrein, was one of the first terpenoids for which a chemical structure was deduced (Lederer et al. 1946; Ruzicka and Lardon 1946). *In vitro*, ambrein is produced from squalene or squalene oxide, by two terminal cyclisations, mediated by two bacterial enzymes, the squalene-hopene cyclase AacSHC mutant D377C and a tetraprenyl-β-curcumene cyclase isolated from *Bacillus megaterium* (Ueda et al. 2013).

The $^{13}C/^{12}C$ isotopic value of the ambergris samples in the present study (Table 1; mean -21.6 ‰) was not statistically different (P=0.001) to that of squalene from sharks (-20.6 ‰; Camin et al. 2010), and similar to values reported for dentine from sperm whales (-14 to -11 ‰; Borell et al. 2013; Mendes et al. 2007)). Such data are thus consistent with a proposed origin from marine organic matter such as squalene, in the whale rectum (Clarke 2006). A bacterially-mediated mechanism is presumably responsible. A bacterium, *Spirillum recti physeteris*, was isolated from an ambergris sample stored for four years (Beauregard 1898), though nothing detailed is known.

The microbiomes of dwarf (*Kogia sima*) and pygmy (*Kogia breviceps*) sperm whales have now been elucidated (Erwin et al. 2017), but not that of the sperm whale. Clarke (2006) stated that “there is reason to believe that [ambergris] also occurs in the pygmy sperm whale” and reviewed the evidence. The gut microbiome of *K. breviceps* is dominated by Firmicutes and Bacteroidetes bacteria. Other core members of kogiid gut biomes were affiliated with sulfate-reducing bacteria (Erwin et al. 2017). This may also be true of the sperm whale, since sperm and pygmy sperm whales have similar cephalopod diets (Clarke 1954; 2006). If so, such microbes might be amongst those involved in the conversion of squalene to ambrein in the sperm whale. Our isotope data (Table 1) certainly do not contradict the likelihood that ambrein is biosynthesised by bacteria from squalene in the whale.

The results of the present study show that, in ambergris, ambrein certainly occurs as one of the major organic-soluble constituents (Table 1), of what are sometimes recorded as huge boulders weighing as much as 455 kg (Clarke 1954; 2006). On the death and decomposition of the whale, ambergris is presumed to be released into the oceans (Clarke 2006). Although numerous studies of the fate of carcasses of sperm whales (whale falls), including video records, have been made, these have not included reports of the fate of ambergris. Nonetheless, our results show that in large coproliths of up to 20 kg (Table 1), ambrein is rather resistant to extensive microbial or photodegradation, under the conditions prevailing, even for periods of about a thousand years (Table 1). This is far longer than had been thought likely previously (Kemp 2018).

3. Conclusions

The samples from Chile studied herein were produced by a whale or whales living 300-500 y before Ferdinand Magellan visited Chile, when the island (Chiloé meaning ‘seagull-land’) was inhabited only by the ancient Chunos people. At this time, in the UK, *Magna carta* had not been signed. This piece of ambergris was thus formed at the time the earliest records of the use of ambergris by humankind were made (Levey 1961). This substantial age indicates that marine preservation of ambergris is more likely than considered hitherto and perhaps lends support to reports that rare lithification of ambergris is represented in the marine geological record (Baldanza et al. 2013; Monaco et al. 2014).

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References

Azzolini M. 2017. Talking of animals: whales, ambergris, and the circulation of knowledge in seventeenth-century Rome. Renaissance Studies 31: 297-318.

Baldanza A. Bizzarri R. Famiani F. Monaco P. Pelligrino R. Sassi P. 2013. Enigmatic,biogenically induced structures in Pleistocene marine deposits: a first record of fossil ambergris Geology 41: 1075-1078.

Baynes-Cope AD. 1962. Analyses of samples of ambergris. Nature 193: 978-979.

Beauregard H. 1898. Les cryptogames de l’ambregris. Annales de Micrographie Paris 10: 241-278.

Berzin AA. 1971. Cachalot. Pacific Scientific Research. Institute of Fisheries and Oceanography, Moscow. pp. 368. (In Russian).

Borrell A. Velásquez Vacca A. Pinela A.M. Kinze C. Lockyer CH. Vighi M. Aguilar A. 2013. Stable isotopes provide insight into population structure and segregation in eastern North Atlantic sperm whales. PLoS ONE 8, e82398.

Boylston Z. 1724. XI. Ambergris found in whales. Communicated by Dr Boylston of Boston in New-England. Philosophical Transactions of the Royal Society 33:193.

Brito C. Jordão VL. Pierce GJ. 2016. Ambergris as an overlooked historical marine resource: its biology and role as a global economic commodity. Journal of the Marine Biological Association of the United Kingdom 96: 585-596.

Camin F. Bontempo L. Ziller L. Piangiolino C. Morchio G. 2010. Stable isotope ratios of carbon and hydrogen to distinguish olive oil and shark squalene-squalane. Rapid Communications in Mass Spectrometry 24:1810-1816.

Clarke R. 1954. A great haul of ambergris. Nature 174: 155-156.

Clarke R. 2006. The origin of ambergris. Latin American Journal of Aquatic Mammals 5: 7-21.

Dugan H. 2011. The ephemeral history of perfume. Scent and sense in modern England. John Hopkins University Press, Baltimore, USA. Chapter 5. Oiled in ambergris.

Erwin PM. Rhodes RG. Kiser KB. Keenan-Bateman T. McLellan WA. Pabst DA.2017. High diversity and unique composition of gut microbiomes in pygmy (Kogia breviceps) and dwarf (K. sima) sperm whales. Scientific Reports 7: 7205.

Kemp C. 2012. Floating gold: a natural (and unnatural) history of ambergris. University of Chicago Press 232 pp.

Kemp C. 2018. Ambergris. Encyclopaedia of marine mammals. (3rd edition) 24-25, Elsevier. (eds: Wűrsig, B., Thewissen, J.G.M. and Kovacs, K.).

Korzh LN. Strigina LI. 1972. Chemical composition of different varieties of ambergris. Maslo-Zhirovaya Promyshlennost 10: 25-26. (In Russian).

Lederer E. Marx F. Mercier D. Pérot, G. 1946. Sur les constituants de l’ambre gris II. Ambréine et coprostanone. Helvetica Chimica Acta 29: 1354-1365.

Levey M. 1961. Ibn Masawaith and his treatise on simple aromatic substances: Studies in the history of Arabic pharmacology I. Journal of the History of Medicine 16: 394-410.

Mendes S. Newton J. Reid RJ. Frantzis A. Pierce GJ. 2007. Stable isotope profiles in sperm whale teeth: variations between areas and sexes. Journal of the Marine Biological Association of the United Kingdom 87: 621-627.

Monaco P. Baldanza A. Bizzarri R. Famiani F. Lezzerini M. Sciuto F. 2014. Ambergris cololites of Pleistocene sperm whales from central Italy and description of the new ichnogenus and ichnospecies Ambergrisichnus alleronae. Palaeontologia Electronica 17: 29A 20pp.

Purchas S. 1613. Purchas his pilgrimage. London. Henrie Fetherstone. 710.

Read S. 2013. Ambergris and early modern languages of scent. *The Seventeenth Century* 28: 221-237.

Reimer PJ. et al. 2013. Intcal13 and marine13 radiocarbon age calibration curves 0–50,000 years cal bp. Radiocarbon 55: 1869-1887.

Rowland SJ. Sutton PA. 2017. Chromatographic and spectral studies of jetsam and archived ambergris. Natural Product Research 31: 1752-1757.

Rowland SJ. Sutton PA. Belt ST. Fitzsimmons-Thoss V. Scarlett AG. 2018. Further spectral and chromatographic studies of ambergris. Natural Product Research [doi: 10.1080/14786419.2018.1428599].

Ruzicka L. Lardon L. 1946. Zur kenntnis der triterpene. (105. Mitteilung). Über das ambreïn, einen bestandteil des grauen ambra. Helvetica Chimica Acta 29: 912.

Schwediawer F-X. Banks J. 1783. XV. An account of ambergrise. *Philosophical Transactions of the Royal Society* 73: 226-241.

Serra S. 2013. An expedient preparation of enantio-enriched ambergris odorants starting from commercial ionone alpha. Flavour and Fragrance Journal 28: 46-52.

Srinivasan TM. 2015. Ambergris in perfumery in the past and present Indian context and the western world. Indian Journal of History of Science 50: 306-323.

Synal HA. Stocker M. Suter M. 2007. MICADAS: A new compact radiocarbon AMS system. Nuclear Instruments and Methods in Physics Research B, 259: 7-13.

Ueda D. Hoshino T. Sato T. 2013. Cyclization of squalene from both termini: identification of an onoceroid synthase and enzymatic synthesis of ambrein. Journal of the American Chemical Society 135: 18335-18338.

Wacker L. Nemec M. Bourquin J. 2010. A revolutionary graphitisation system: Fully automated, compact and simple. Nuclear Instruments and Methods in Physics Research B, 268: 931-934.

Figure 1. Locations of finds of jetsam ambergris samples. Numbers refer to samples listed in Table 1. (The exact location of sample 20 is unknown).

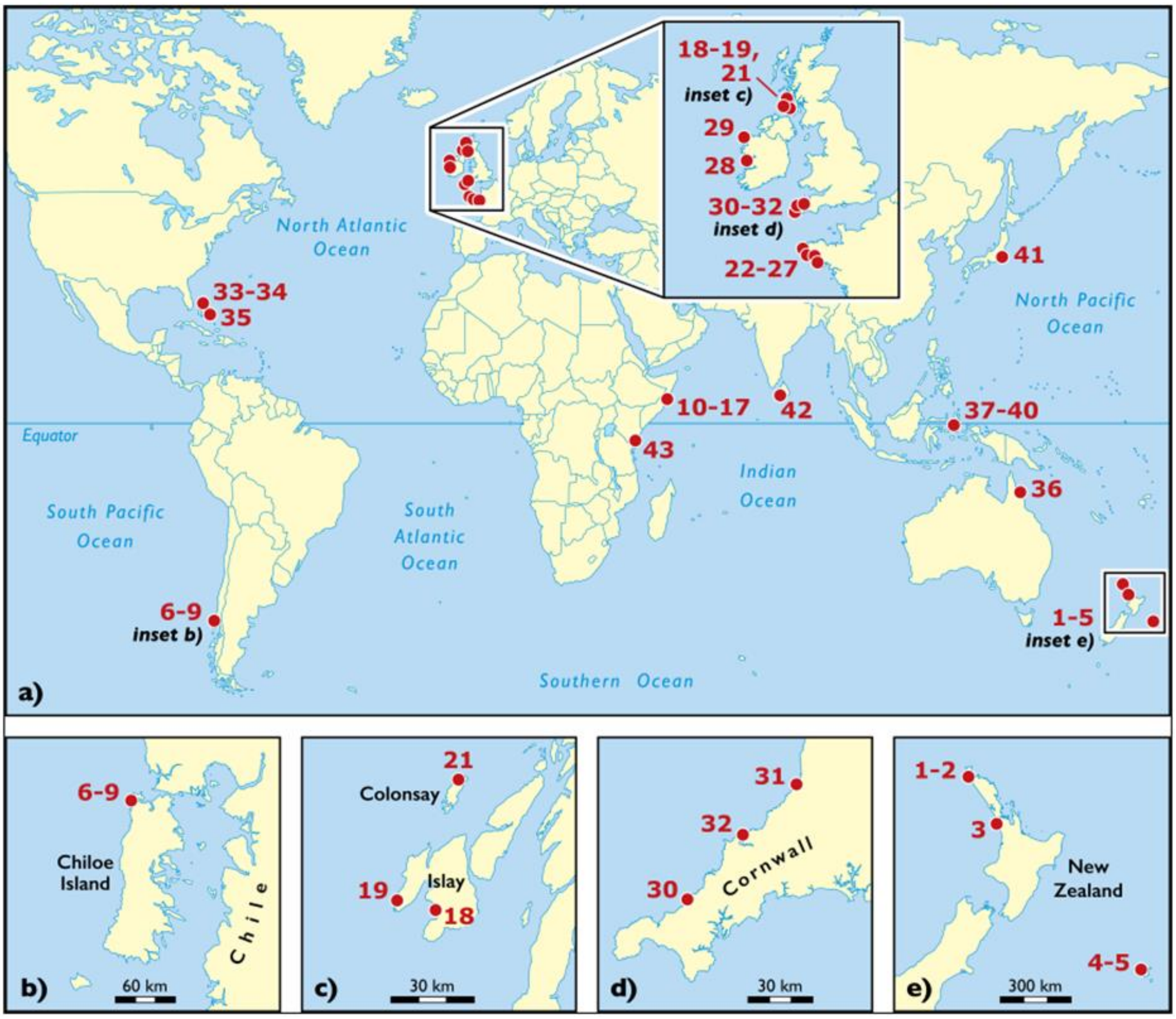

Table 1. Locations, dates and amounts of finds of jetsam ambergris samples with ^{13}C data and ^{14}C isotopic radiocarbon ages. Key: [a]= total weight of unrelated pieces in same collection batch; [b,c] = total weight of pieces in same collection batch [d] = % of ambrein as TMS ether as determined by GC-MS. (The calibration curves for the radiocarbon to calendar age conversions are shown in Fig. S1) [e]= Calibrated age of mean of samples 6-9 (Figure S1) n.d.= not determined.

Code	Beach Location	Country	Total Weight (g)	Stated Collection Date	% DCM extractable	% ambrein [d]	δ13C ‰	^{14}C age (y)	$F^{14}C$	Calibrated age (y)
1	90 mile beach, North Island	New Zealand	2100	03/02/2017	97	77	-23.9	-202	1.026±0.002	
2	90 mile beach, North Island	New Zealand	2600	03/02/2017	97	97	-23.3	290	0.965±0.002	
3	Cochrane's Gap, Awhitu Peninsula, North Island	New Zealand	Unknown	00/06/2017	99	49	n.d.	n.d.		
4	Pitt Island	New Zealand	50	00/12/2017	101	92	n.d.	n.d.		
5	Pitt Island	New Zealand	20	00/12/2017	96	83	n.d.	n.d.		
6	Mar Brava, Chiloe Island	Chile	6300 [a]	12/04/2017	100	97	-20.6	1517	0.828±0.002	1102-1237 [e]
7	Mar Brava, Chiloe Island	Chile	6300 [a]	12/04/2017	100	92	-21.5	1545	0.825±0.002	
8	Mar Brava, Chiloe Island	Chile	6300 [a]	12/04/2017	100	97	-21.3	1541	0.826±0.002	
9	Mar Brava, Chiloe Island	Chile	6300 [a]	12/04/2017	100	97	-21.5	1548	0.825±0.002	
10	Somalia Grand	Somalia	3000 [b]	05/05/2017	92	86	-22.0	545	0.935±0.002	184-328
11	Somalia Grand	Somalia	3000 [b]	05/05/2017	93	92	-23.6	219	0.973±0.002	
12	Somalia Grand	Somalia	3000 [b]	05/05/2017	97	86	-22.0	63	0.992±0.002	
13	Somalia Grand	Somalia	3000 [b]	05/05/2017	97	82	-25.0	-23	1.003±0.002	
14	Somalia Grand	Somalia	3000 [b]	05/05/2017	92	88	-23.3	113	0.986±0.002	
15	Somalia Grand	Somalia	3000 [b]	05/05/2017	98	62	-25.5	-145	1.018±0.002	
16	Somalia Grand	Somalia	3000 [b]	05/05/2017	95	90	-22.7	-90	1.011±0.002	
17	Somalia Grand	Somalia	3000 [b]	05/05/2017	88	68	-23.3	-214	1.027±0.002	
18	The Big strand, Islay	Scotland	94	10/06/2017	102	93	-21.9	-240	1.030±0.002	
19	Lossit Bay, Islay	Scotland	95	10/06/2017	99	99	-21.7	-325	1.041±0.002	
20	Hebrides	Scotland	90	01/04/2016	97	93	-21.5	-42	1.005±0.002	
21	Kiloran Bay, Colonsay	Scotland	82	03/01/2018	96	97	-20.9	-200	1.025±0.002	
22	Plouhinec, Brittany	France	50	01/09/2017	59	90	-21.3	139	0.983±0.002	
23	Plouhinec, Brittany	France	150	01/09/2017	98	92	-23.2	-55	1.007±0.002	
24	Plouhinec, Brittany	France	180	01/09/2017	100	98	-20.5	-210	1.027±0.002	
25	Plouhinec, Brittany	France	30	01/09/2017	104	74	-22.9	-447	1.057±0.002	
26	Beach of Bretagne	France	2733	20/12/2017	93	97	n.d.	n.d.		
27	Plouhinec, Brittany	France	<1	01/09/2017	101	85	-21.2	8	0.999±0.002	
28	County Clare	Ireland	4	31/07/2017	96	66	-20.7	-65	1.008±0.002	
29	County Mayo	Ireland	88	10/12/2014	97	93	-17.1	110	0.987±0.002	
30	Porthtowan Beach, Cornwall	England	110	01/01/2014	100	92	-21.8	-210	1.027±0.002	
31	Widemouth Bay, Cornwall	England	52	04/03/2018	100	91	n.d.	n.d.		
32	Polzeath, Cornwall	England	13	28/04/2108	100	96	n.d.	n.d.		
33	Aboco	Bahamas	Unknown	Unknown	99	98	-15.9	-139	1.018±0.002	
34	Aboco	Bahamas	Unknown	Unknown	99	85	-21.4	-60	1.008±0.002	
35	Cat Island	Bahamas	826	01/03/2018	97	91	n.d.	n.d.		
36	Cooktown, Queensland	Australia	20000	00/00/2016	103	84	n.d.	n.d.		
37	Bacan island	Indonesia	17000 [c]	00/00/1939	88	76	n.d.	n.d.		
38	Bacan island	Indonesia	17000 [c]	00/001939	97	5	n.d.	n.d.		
39	Bacan island	Indonesia	17000 [c]	00/00/1939	97	32	n.d.	n.d.		
40	Bacan island	Indonesia	17000 [c]	00/00/1939	99	10	n.d.	n.d.		
41	Japan	Japan	300	00/00/2017	87	59	n.d.	n.d.		
42	West Sri Lanka	Sri Lanka	101	16/03/2018	100	60	n.d.	n.d.		
43	Mombasa beach	Kenya	75	00/11/2017	100	97	n.d.	n.d.		

SUPPLEMENTARY MATERIAL

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Abstract

Ambergris, which is a coprolith originating from the sperm whale, has been found only rarely, but for centuries, as jetsam on beaches all over the world. There are no reliable data indicating how long such samples may have remained at sea, with unsubstantiated accounts suggesting maybe decades. Here, we obtained over forty jetsam samples collected many on known dates, from mostly known beach locations across the globe. Such an inventory of verified jetsam ambergris is unprecedented. Each sample was characterised by analytical methods such as gas chromatography-mass spectrometry (GC-MS). We then determined the radiocarbon ages of some of the samples by well-described accelerator-MS techniques. Surprisingly, some samples of jetsam have remained in the environment for about a thousand years.

Experimental

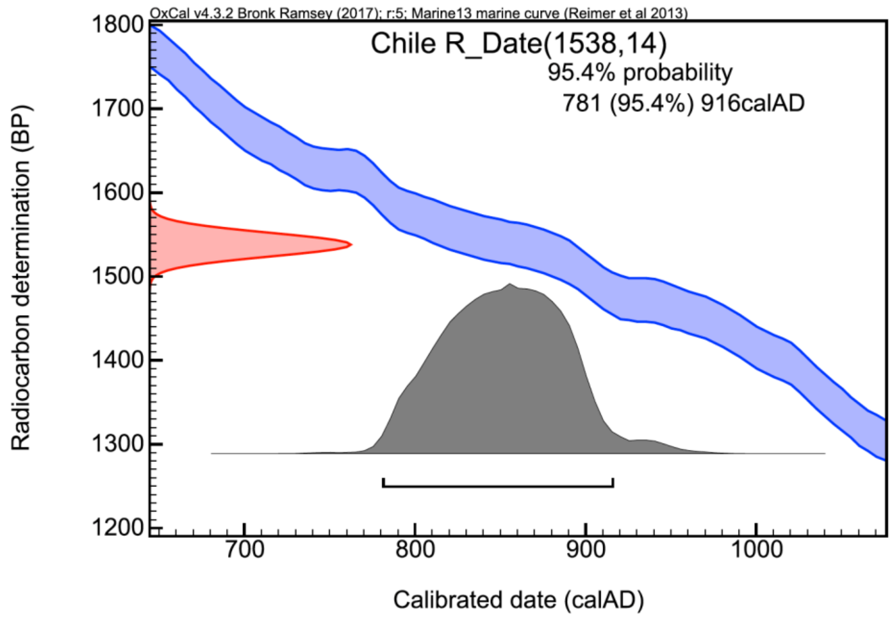
Materials

Jetsam ambergris samples were collected from beaches worldwide, including by ourselves, and on receipt were stored in a dry dark cabinet prior to analysis (Rowland and Sutton 2017).

Methods

The methods for GC-MS analysis have been published (Rowland and Sutton 2017). Methods for AMS are well described (Bronk Ramsey 2017). Due to their chemical purity (mean 81% ambrein; Table 1), samples were graphitised (Wacker et al. 2010) without pretreatment and analysed using a MICADAS AMS (Synal et al. 2007).

Figure S1. Example calibration curves for samples from Chile (samples 6-9) and Somalia (sample 10).



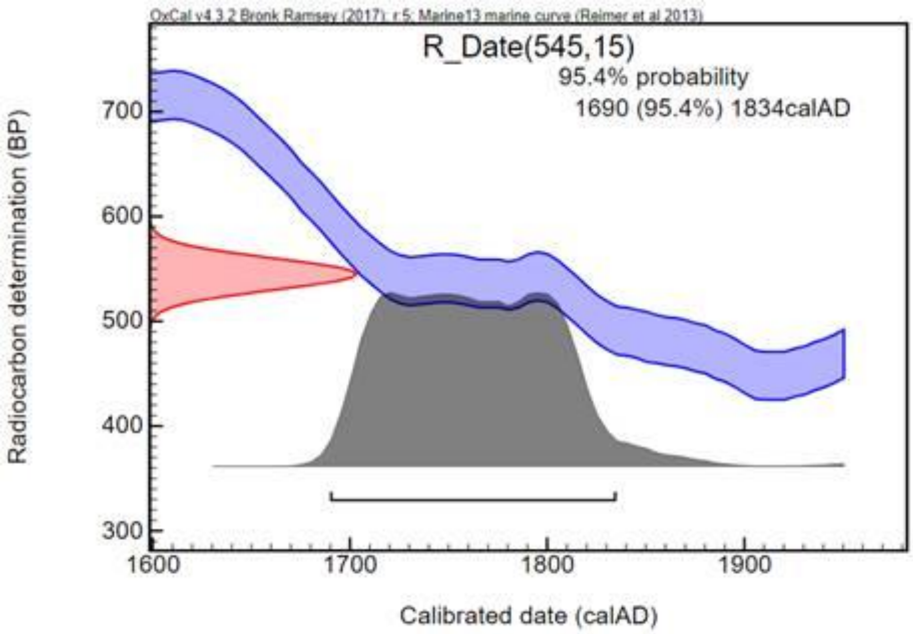


Figure S2. Calibration curve for an additional sample from Mar Brava, Chiloé, Chile

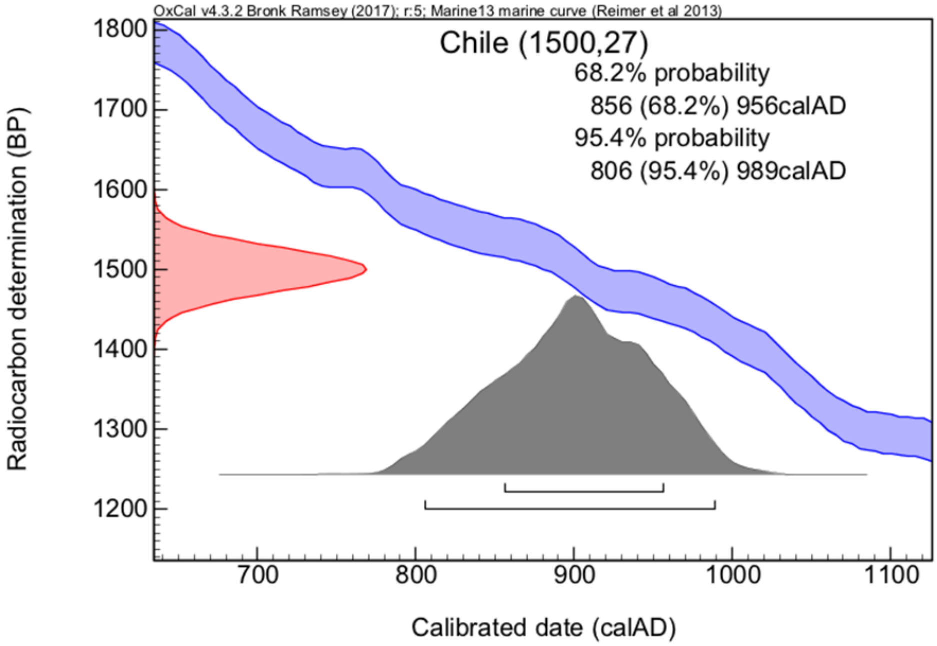


Table S1. Radiocarbon and ^{13}C data for ambergris collected from a dead sperm whale beached on 15 December 2012 at Razende Bol near Texel, Netherlands and archived by the Ecomare Museum, Texel and an additional sample from Chile (calibration for the latter is shown in Figure S2).

Sample Name	**$\delta^{13}C$ (‰)**	**^{14}C Age (BP)**	**$F^{14}C$**
Texel 15.12.12	-21.1	-486	1.062±0.003
Mar Brava Chiloé Chile	-21.1	1500	0.829±0.003

References for Supplementary Information

Bronk Ramsey C. 2017. Methods for Summarizing Radiocarbon Datasets. Radiocarbon, 59: 1809-1833.

Reimer P.J. et al. 2013. Intcal13 and marine13 radiocarbon age calibration curves 0–50,000 years cal bp. Radiocarbon, 55: 1869-1887.

Rowland SJ. Sutton PA. 2017. Chromatographic and spectral studies of jetsam and archived ambergris. Natural Product Research 31: 1752-1757.

Wacker L. Nemec M. Bourquin J. 2010. A revolutionary graphitisation system: Fully automated, compact and simple. Nuclear Instruments and Methods in Physics Research B, 268: 931-934.