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Rapid Early Development of Circumarctic Peatlands and Atmospheric CH_4 and CO_2 Variations

Glen M. MacDonald,[1,2]* David W. Beilman,[1] Konstantine V. Kremenetski,[1,3] Yongwei Sheng,[1] Laurence C. Smith,[1] Andrei A. Velichko[3]

An analysis of 1516 radiocarbon dates demonstrates that the development of the current circumarctic peatlands began ~16.5 thousand years ago (ka) and expanded explosively between 12 and 8 ka in concert with high summer insolation and increasing temperatures. Their rapid development contributed to the sustained peak in CH_4 and modest decline of CO_2 during the early Holocene and likely contributed to CH_4 and CO_2 fluctuations during earlier interglacial and interstadial transitions. Given the decreased tempo of peatland initiation in the late Holocene and the transition of many from fens (which generated high levels of CH_4) to ombrotrophic bogs, a neoglacial expansion of northern peatlands cannot explain the increase in atmospheric CH_4 that occurred after 6 ka.

Modern northern peatlands cover about 4 million km² across Eurasia and North America and represent the biggest wetland complex in the world (Fig. 1). Today, these peatlands are thought to store 180 to 455 Pg of sequestered carbon while also releasing 20 to 45 Tg per year of CH_4 into the atmosphere (*1*, *2*). The potential contribution of northern peatlands to fluctuations in atmospheric CH_4 and CO_2 over the late glacial and Holocene, and during earlier interglacials, has been a matter of much speculation and debate (*3*–*8*).

Ice-core records show that CH_4 concentrations rose from ~350 to 650 parts per billion by volume (ppbv) between the last glacial maximum (LGM), which occurred 20 ka (20,000 calendar years before C.E. 1950), and the Bølling-Allerød (BA) warm period (~15 to 13 ka). They then declined by ~200 ppbv during the Younger Dryas (YD) stadial (~13 to 11.5 ka), rose rapidly to levels over 700 ppbv in the early Holocene (11 to 8 ka), and then declined again between 8 and 6 ka (*3*). It has been maintained that because conditions were not favorable for widespread circumarctic peatland formation until after 8 ka, tropical wetlands or marine clathrates were the likely sources for the CH_4 peak that occurred 11 to 8 ka (*4*, *9*). On the basis of the assumed late-Holocene development, it has been suggested that northern peatlands played little role in the declining atmospheric CO_2, which has also been observed during the period from 11 to 8 ka (*5*). Others argue that the development of the northern peatland complex contributed substantially to the early-Holocene CH_4 increase and simultaneously decreased atmospheric CO_2 through carbon sequestration in northern soils (*6*–*8*).

Resolving the debate on the potential role of the northern peatlands in early postglacial CH_4 variations has become critical since the recent analysis of the deuterium and carbon isotopic composition of CH_4 (δD_{CH_4} and $\delta^{13}C_{CH_4}$) from Greenland ice samples, which suggested that the destabilization of marine clathrates is an unlikely explanation for the BA or early-Holocene CH_4 increases (*10*, *11*). In view of this evidence, it has been argued that the sustained high levels of CH_4 that developed at the close of the YD in part require a persistent terrestrial source linked to the warming climate at that time (*11*).

Holocene concentrations of atmospheric CH_4 reached a minimum of <600 ppbv at 6 ka and then increased again over the late Holocene to values of about 695 ppbv just before the industrial revolution (*3*). This late-Holocene increase has been variously attributed either to expansion of northern wetlands due to neoglacial climatic cooling after the Holocene thermal maximum (*4*) or to the product of expanding anthropogenic activity (particularly the expansion of rice- and cattle-based agrarian societies) in the mid- to late Holocene (*12*). However, recently collected CH_4 data from Antarctic ice cores reveal that the mid- to late-Holocene increase is not unique. A similar late-interglacial increase in Pleistocene atmospheric CH_4 occurred ~400 ka during Marine Isotope Stage 11 (MIS11), which clearly cannot reflect anthropogenic sources and has been ascribed instead to natural factors, including expansion of northern wetlands (*13*).

To address the hypothesis that northern peatland development could have contributed to the late-Pleistocene and Holocene variations in atmospheric CH_4 and CO_2 outlined above, we collated 1516 basal radiocarbon dates for peat initiation from wetlands throughout high-latitude Europe, Asia, and North America from a wide variety of sources (*14*). Some areas, such as Fennoscandia, have numerous basal dates for a small geographic area, whereas other very large areas such as central and eastern Siberia have a limited number of dates (Fig. 1). Therefore, we analyzed the compiled data set by raw number of initiation dates, and we also divided the Northern Hemisphere into grids of 2° latitude by 2° longitude and assigned a value for peatland initiation based on the oldest basal radiocarbon date in each cell (Fig. 1).

The lack of basal dates older than about 16.5 ka suggests that there was no extensive peatland complex in the northern circumpolar region during the LGM (Fig. 2). This finding is corroborated by palynological data that indicate a paucity of *Sphagnum* (peat moss) spores from deposits of this age (*15*). Before 16.5 ka, much of the North American and European arctic and subarctic were still covered in ice, and it is likely that the large ice-free areas of Siberia and Beringia were too cold and dry (*16*) to promote extensive peatland development. This absence of any significant northern peatland complex during the LGM is consistent with the depressed CH_4 levels and the relatively low proportion of northern CH_4 sources observed in ice-core records (Fig. 3).

In concert with increasing summer insolation and northern high-latitude temperatures, the current northern peatland complex began developing in ice-free portions of North America and Asia between 16.5 and 14 ka and initiating widely on all three northern continents after 14 ka (Figs. 1 and 2). These results dispel the earlier assertion that peatland development in

[1]Department of Geography, [2]Department of Ecology and Evolutionary Biology, University of California, Los Angeles, CA 90095–1524, USA. [3]Russian Academy of Sciences, Moscow 109017, Russia.

*To whom correspondence should be addressed. E-mail: macdonal@geog.ucla.edu

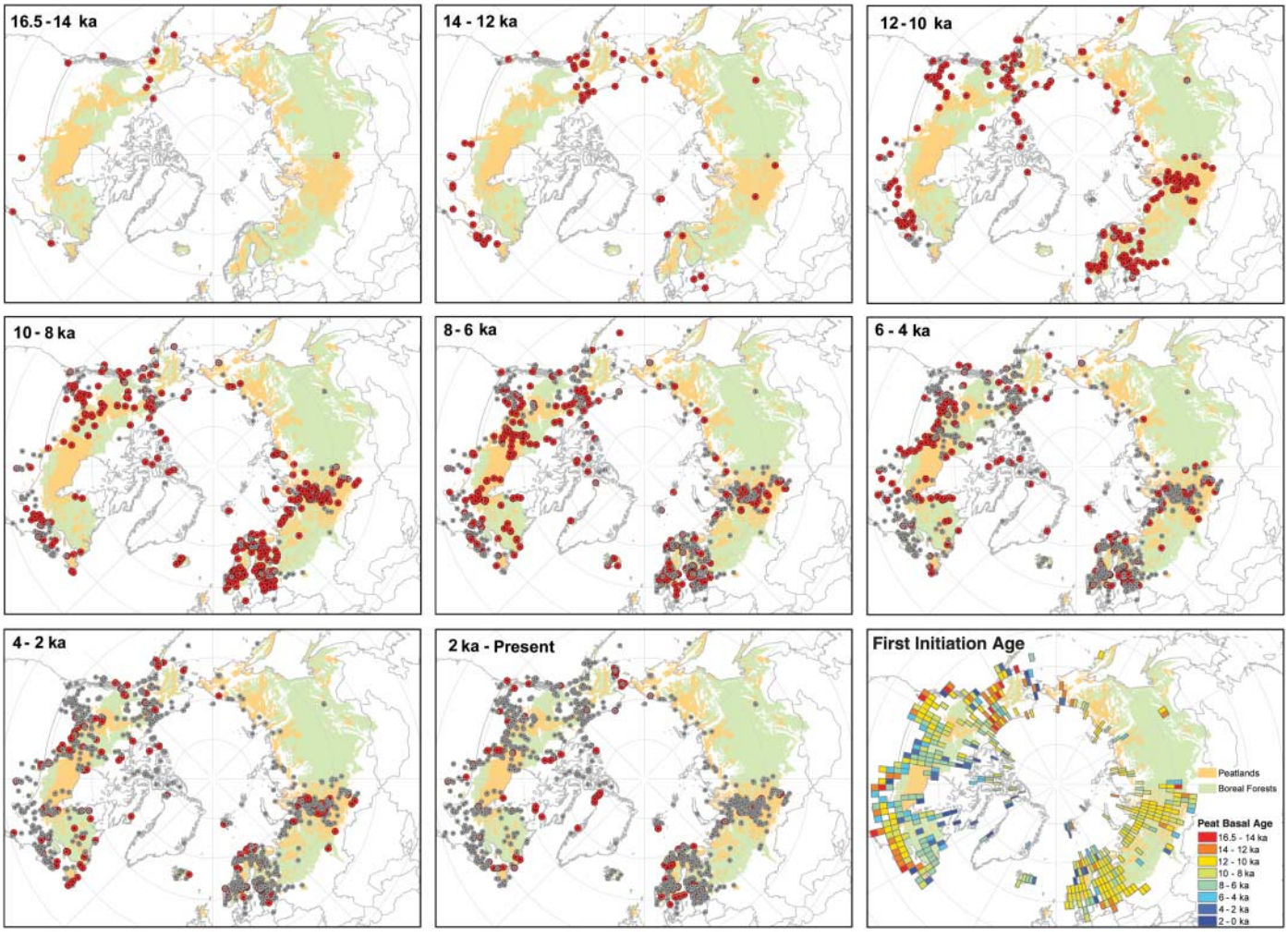

Fig. 1. Map of the current distribution of the northern circumarctic peatland complex and the boreal forest biome developed from a number of sources (SOM text) and the initiation dates of the peatlands based on radiocarbon dates from the base of peat deposits (*14*). The red dots indicate new peatlands that initiated during each time slice and the gray dots indicate preexisting peatlands initiated during earlier time slices. The timing of peatland initiation based on 2° by 2° grids is shown in the right panel in the bottom row. The grids map the oldest basal peat dates within each grid cell.

western Siberia was in advance of development in North America or elsewhere (*7*). The initial expansion coincides with warming during the BA period (Fig. 3), and it is likely that increasing warmth and moisture and decreasing glacial ice cover promoted peatland growth. Resultant development of the northern peatland complex corresponds with increasing atmospheric CH_4 concentrations observed during the BA (Fig. 3). However, the still-limited extent of northern peatlands at this early interval is consistent with evidence for a relatively restricted role of northern wetlands in producing the BA CH_4 increase (Fig. 3). There is a small decline in the rate of new peatland formation during the YD, which may be attributed to the development of cold conditions over much of the Northern Hemisphere and readvances of ice. At the same time, there is a precipitous drop in CH_4 recorded in the ice cores (Fig. 3) that seems to reflect additional factors. During the BA and YD periods, it is unlikely that the extent and growth of the northern peatlands was great enough to have significantly affected atmospheric CO_2 concentrations.

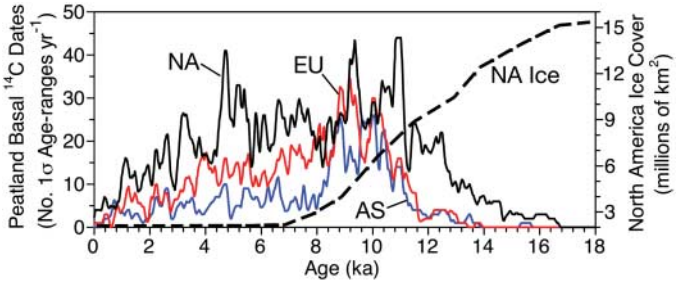

Fig. 2. Timing of circumarctic peatland establishment in North America (black), Europe (red), and Asia (blue) based on the total number of initiation dates from each region. The occurrence frequency of basal peat radiocarbon ages is plotted as the number of calibrated age ranges that fall in any year (*14*), smoothed with a 100-year running mean. The decreasing area of the Laurentide Ice Sheet (dashed line) as it retreated over the late glacial and Holocene (*17*) is also plotted.

The initiation of the early Holocene warming at 11.5 ka after the YD is marked by rapid expansion of peatlands throughout the north. The North American peatlands expanded rapidly during this time (Figs. 1 and 2), even though still constrained by the ice extent of the Laurentide Ice Sheet (*17*). The tempo of subsequent northern peatland development in North America was influenced by the rate of continued ice retreat and the exposure of land surfaces (Fig. 2). Large areas of northern Asia never supported ice during the LGM (*18*), and by 11 ka, high-latitude climate had warmed (Fig. 3). What remained of the Scandinavian Ice Sheet was restricted to cen-

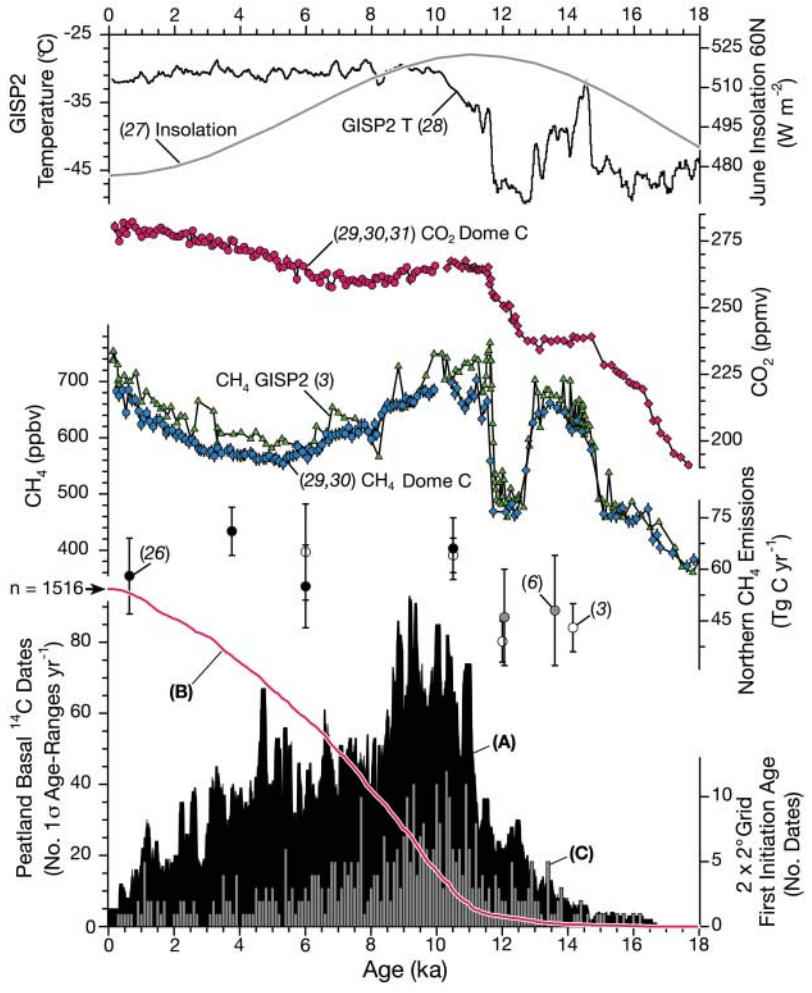

Fig. 3. Timing of circumarctic peatland establishment compared with June insolation at 60°N (*26*), Greenland Ice Sheet Project 2 (GISP2) temperature reconstruction (*27*), atmospheric CO_2 and CH_4 concentrations, and estimates of Northern Hemisphere CH_4 emissions derived from the InterPolar CH_4 Gradient (IPG) (*3*, *4*, *6*, *28*). Atmospheric CO_2 and CH_4 concentrations show ice-core data from European Project for Ice Coring in Antarctica (EPICA) Dome C (red for CO_2 and blue for CH_4) and GISP2 [green triangles show CH_4 (*3*, *29*–*31*)]. Dome C data are shown on the original EPICA Dome C 1 time scale from 0 to 10 ka before present (circles) and on the GISP2 time scale with CH_4 synchronization from 10 to 18 ka before present (diamonds; SOM text). Dome C error bars indicate 1σ uncertainty. (**A**) The occurrence frequency of 1516 radiocarbon dates of basal peat deposits (*14*) shows the number of calibrated age ranges that occur in any year (black curve). (**B**) Cumulative curve of 1516 dates (red curve). (**C**) The oldest basal peat dates within each 2° by 2° grid (gray bars).

tral Fennoscandia (*19*), allowing widespread expansion of peatlands in northern Eurasia (Figs. 1 and 2). A sustained period of maximum rates of peatland establishment followed and persisted until about 8 ka.

The rapid expansion of peatlands at the close of the YD coincides with a ~15-Tg increase in atmospheric CH_4 derived from the Northern Hemisphere (Fig. 3) and a 200- to 250-ppbv increase in total CH_4 concentrations. According to our data, the northern peatland complex was likely at <20% of its current aerial extent at the end of the YD and expanded to about 50% by 8 ka. On the basis of current estimates of overall CH_4 production from northern peatlands (*1*, *2*), they may have contributed 4 to 9 Tg of Northern Hemisphere CH_4 after the end of the YD and up to 12 to 27 Tg by 8 ka. However, based on typical peatland succession stages, higher summer insolation in the early Holocene, and evidence of peatland vegetation and type from our peat cores taken in Siberia (*7*), it is likely that many of these newly developed peatlands were warm and wet minerotrophic fens, often dominated by sedges. Such fens typically emit CH_4 at rates many times greater than the ombrotrophic *Sphagnum* bogs common in much of the north today (*6*, *20*–*22*). Therefore, we suspect that the rates of CH_4 production in northern peatlands may have been considerably higher in the early Holocene than they are today.

The $\delta^{13}C_{CH_4}$ values of ice from the Pakitsoq outcrop in western Greenland have been used to infer the likely origins of early-Holocene atmospheric CH_4 (*11*). It has been suggested that major terrestrial sources contributing methane during the period from 11 to 8 ka likely produced CH_4 with $\delta^{13}C$ values of between –50 and –60 (*11*). Although northern peatlands dominated by ombrogenous bogs may typically emit CH_4 with $\delta^{13}C < -60$, pore waters and emissions of boreal fens are relatively enriched in ^{13}C (average $\delta^{13}C_{CH_4}$ values of –50 to –60), particularly in high-productivity sites (*23*). These values are consistent with the early northern peatland complex, dominated by minerotrophic fens, which was a major contributor to the peak in CH_4 that occurred 11 to 8 ka and must be considered in addition to the potential tropical sources suggested previously (*11*).

The rapid growth of the circumarctic peatland complex and its associated sequestration of phytomass carbon in the early Holocene may have also contributed to the decline by ~7 parts per million by volume (ppmv) of atmospheric CO_2 observed in the ice-core records between 11 ka and the mid-Holocene (Fig. 3). This decline has been interpreted as reflecting a total biosphere uptake of 110 Pg C in the first half of the Holocene (*8*). More than half of the peatland basal dates in our data set are older than 8 ka, indicating rapid initiation and development of this carbon sink in the early Holocene. Conservatively, if by 8 ka peat deposits were 0.5 to 1 m thick, covered just one-quarter of today's peatland area, and were similar in carbon characteristics to today's northern peatlands (*1*), they would have been capable of sequestering 29 to 58 Pg C. Further detailed reconstruction of net peat accumulation rates during this period are required to better estimate the magnitude of the contribution of the northern peatlands to early-Holocene atmospheric CO_2 declines.

The widespread development of peatlands in response to increasing summer insolation, BA warming, and particularly rapid Holocene warming after the YD supports the hypothesis that the northern peatlands were a major terrestrial factor contributing to the early fluctuations in atmospheric CH_4 and to CO_2 sequestration during the current interglacial. It is likely that they played a similar role in earlier interglacials. The rapidity and large spatial extent of the response of the northern peatland complex, particularly to the onset of post-YD warming, suggests that they may even have played a role in CH_4 and CO_2 variations at the shorter time scale of earlier interstadials such as MIS3.

The observed decline in peatland initiation, particularly in Europe and Asia after 8 ka, corresponds to a decline in atmospheric CH_4 concentrations between 8 and 6 ka (Fig. 3). This decline in initiation rates should not be confused with a decline in total peatland area; most peatlands that were extant at 8 ka were still extant at 6 ka. However, many had trans-

formed from early minerotrophic fens to ombrotrophic *Sphagnum* bogs, which are typically weaker sources of CH_4 than are fens (*20*–*23*). Therefore, the transformation from high CH_4-efflux fens to *Sphagnum* bogs, coupled with a declining rate of new peatland formation, would have contributed to the decline in atmospheric CH_4. What remains puzzling is the role that northern peatlands played in the subsequent increase in CH_4 between 6 ka and just before the Industrial Revolution. By C.E. 1700, levels of atmospheric CH_4 had increased once again to almost 700 ppbv. However, contrary to earlier speculation (*9*, *15*), new peatland initiation was relatively modest in the late Holocene, and conversion of fens producing high levels of CH_4 to *Sphagnum* bogs with lower production was ongoing. If the mid- to late-Holocene CH_4 increase does not have an anthropogenic explanation, then its source must lie in factors other than large-scale resurgent expansion of the northern peatland complex.

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Supporting Online Material

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Table S1
References

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Gold in Magmatic Hydrothermal Solutions and the Rapid Formation of a Giant Ore Deposit

Stuart F. Simmons[1]* and Kevin L. Brown[2]

The Ladolam hydrothermal system, on Lihir Island, Papua New Guinea, hosts one of the youngest and largest gold deposits in the world. Several deep (more than 1 kilometer) geothermal wells were drilled beneath the ore bodies to extract water at >275°C and to facilitate open-pit mining. Using a titanium down-hole sampler, we determined that the deep geothermal brine of magmatic origin contains ~15 parts per billion gold. At the current gold flux of 24 kilograms per year, this deposit could have formed within ~55,000 years. The combination of sustained metal flux and efficient metal precipitation led to the formation of a giant hydrothermal gold deposit in a short period.

The origins of giant hydrothermal gold deposits are enigmatic (*1*). This is because the concentrations of precious metals and flow rates of ore-forming fluids are poorly quantified, and the origins of the metals are unclear. These aspects can be clarified with direct analyses of fluids from modern hydrothermal systems. The only known active hydrothermal gold deposit is at Ladolam, on Lihir Island, Papua New Guinea, and this deposit is one of the largest in the world with ~1300 tons of gold. Geothermal drilling at Ladolam has been ongoing since before the start of open-pit mining in 1997, and the wells provide access to the deep fluids upstream of the ore zone. Because precious metals precipitate in the wells during fluid ascent due to boiling (*2*), we used a titanium down-hole sampler to obtain deep fluids for analyses (*3*). Any metals that precipitate in the sampler during its return to the surface can be incorporated into the water sample by rinsing the titanium sampler with strong acid. Thus, the deep metal concentrations can be directly measured and used to constrain the rate at which the gold is transported and then deposited in order to form this giant deposit. Because the isotopic compositions of the deep hydrothermal waters at Ladolam are predominantly magmatic in origin (*4*), our data cover not just the origin of the metals, but also the concentrations of gold and related metals in magmatic hydrothermal solutions, which are thought to be important to the formation of ore deposits (*5*, *6*).

The Ladolam gold deposit occupies the center of the extinct Luise volcano on Lihir Island (Fig. 1). The two major tabular ore zones cover ~2 km^2 and extend from the surface to 400 m below sea level. They lie in the middle of a breached crater that formed in response to sector collapse and unroofing of the volcanic edifice ~400,000 years ago (*4*, *7*, *8*). The resulting explosive depressurization of the magmatic-hydrothermal system produced a diatreme breccia complex and highly permeable rocks, which now host the ore. Mineralogical, fluid inclusion, and isotope studies (*4*) show that the gold was deposited in two stages between 150° and 250°C, from solutions of magmatic origin, when they mixed with other fluids or boiled. The magmatic gold-bearing solutions were near neutral to slightly alkaline pH and contained 5 to 10 wt % equivalent NaCl. The host rocks comprise alkalic mafic to intermediate volcanic and intrusive rocks (*9*) that have been hydro-

[1]Geology Dept., Univ. of Auckland, Private Bag 92019, Auckland Mail Centre, Auckland 1142, New Zealand. [2]GEOKEM, P. O. Box 95-210, Swanson, Waitakere 0653, New Zealand.

*To whom correspondence should be addressed. E-mail: sf.simmons@auckland.ac.nz