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Evidence for natural molecular hydrogen seepage associated with Carolina bays (surficial, ovoid depressions on the Atlantic Coastal Plain, Province of the USA)

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Abstract

A study of soil gases was made in North Carolina (USA) in and around morphological depressions called "Carolina bays." This type of depression is observed over the Atlantic coastal plains of the USA, but their origin remains debated. Significant concentrations of molecular hydrogen (H_2) were detected, notably around the bays. These measurements suggest that Carolina bays are the surficial expression of fluid flow pathways for hydrogen gas moving from depth to the surface. The potential mechanisms of H_2 production and transport and the geological controls on the fluid migration pathways are discussed, with reference to the hypothesis that Carolina bays are the result of local collapses caused by the alteration of rock along the deep pathways of H_2 migrating towards the surface. The present H_2 seepages are comparable to those in similar structures previously observed in the East European craton.

Keywords: Hydrogen; Seepage; Carolina bays

Background

Carolina bays are surficial, consistently oriented, oval-shaped depressions that occur widely across the southeastern Atlantic Coastal Plain, Province of eastern North America (Brooks et al. 2010). They are well defined on satellite images (Figs. 2, 3, and 4) and densely cover parts of the Coastal Plain in North Carolina (NC) and South Carolina (SC). They vary in size, ranging from ~100 m to 8 km in diameter (Lake Waccamaw, NC, USA). Slightly elevated rims (~1–3 m), commonly consisting of sand, surround these features. Although some bays have continuous elevated rims, the rims do not usually completely encircle the bays but often form a crescent. The long axes of these elliptical features are preferentially oriented NW–SE (Figs. 1, 2, 3, and 4). Bays of various sizes may overlap, and small bays are frequently present inside larger bays (Fig. 2). In areas undisturbed by anthropogenic activities, these bays can include densely vegetated wetlands or open water lakes. These features were originally called "bays" because of the bay trees that inhabit these wet depressions or pocosins. Now, the term "bay" indicates a wet oval-shaped depression. Locally, these features are also called cypress domes, Grady ponds, citronelle ponds, wet prairies, sandhill ponds, etc. (Folkerts 1997). In anthropogenically modified areas, the bays are commonly drained and cleared for agriculture or other purposes. Even when modified, most of the bays are still easily discernable in satellite and Light Detection and Ranging (LiDAR) images because of their characteristic morphology and relief and the soil bleaching on their rims. Hundreds of thousands of bays occur along the Atlantic Coastal Plain from New Jersey to Florida (Prouty 1952), and in NC, the bays cover as much as 65 % of the land surface of the Coastal Plain (Prouty 1952). Eyton and Parkhurst (1975) summarized the physical characteristics of Carolina bays.

The origin of Carolina bays is still unclear. Many theories have been proposed to explain their formation,

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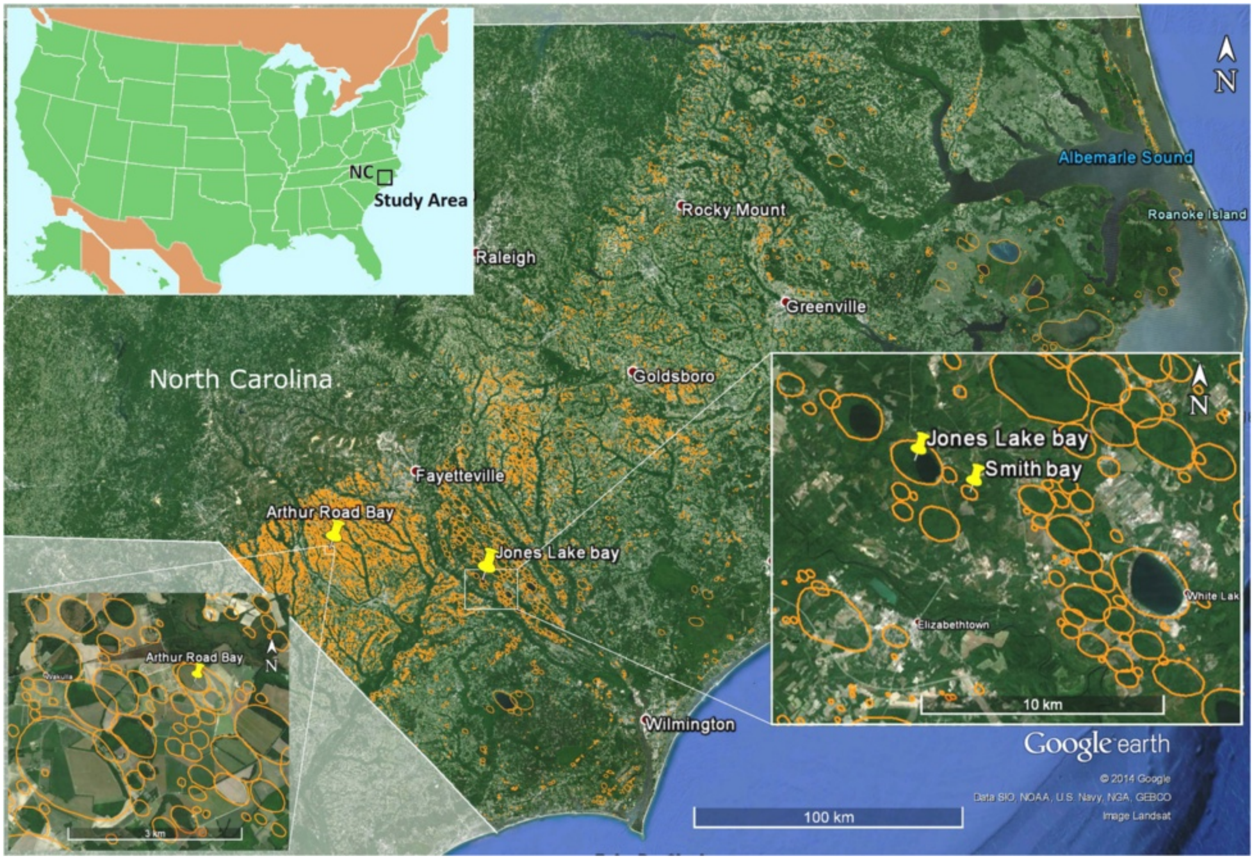

Fig. 1 Location of the study area. *NC* North Carolina. Carolina bays are outlined by *orange polygons*. This and all other satellite images in this article were downloaded from Google Earth and Google Earth Pro

including meteorite impact, comet impact, wind action, water table fluctuations, solution phenomena, and even fish nests (Ross 1987), but no general consensus has yet been reached.

In the present study, we examined the chemical compositions of the gases from the soils in selected Carolina bays and their vicinities to investigate the contents of H_2 and other reduced gases. Our aim was to compare the bays with similar geomorphological features recently described in the East European craton (EEC) in Russia, which were shown to emit H_2 gas in an area where the occurrence of H_2 at depth (several hundred meters) has been confirmed in boreholes (Larin et al. 2014). A variety of mechanisms can produce H_2, and there may be a causal link between these H_2 emissions and the formation of these structures. The possible relationships between the H_2 seepages and the observed geomorphological features were investigated.

Geological framework

The Atlantic Coastal Plain, Province of eastern North America, is the emergent part of a platform on a passive continental margin and is currently characterized by incised valleys that separate flat interfluves (Farrell et al. 2013). The continental shelf is underlain by a seaward-thickening wedge of sediment that thins westward to a feather edge along its boundary with the crystalline bedrock. This clastic wedge attains a maximum thickness of 3009 m (9854 ft) on land at Cape Hatteras (Lawrence and Hoffman 1993) and includes deposits of Cretaceous to Quaternary age (Brown et al. 1972). The sediment composition and recurring sedimentary facies on the NC Coastal Plain have been summarized by Farrell et al. (2012, 2013).

The NC Coastal Plain is predominantly a relict Plio-Pleistocene landscape, characterized by a series of progressively younger scarps, or paleoshorelines, and intervening terraces that step down in elevation and age towards the coast and towards the river incisions (see Fig. 1 in Farrell et al. 2013 and Figs. 1, 2, 3, and 4 in Abbott et al. 2011), forming a stair-step topography. Over the past 5 Ma, glacio-eustatic changes in sea level drove the transgressive–regressive cycles that sculpted this landscape. The surficial deposits that underlie the relict landscape include a complex assemblage of marine, barrier island, estuarine, fluvial, and other coastal plain deposits, which are predominantly Pliocene, Pleistocene, and Holocene in age. Abbott et al. (2011) summarized the surficial geology and

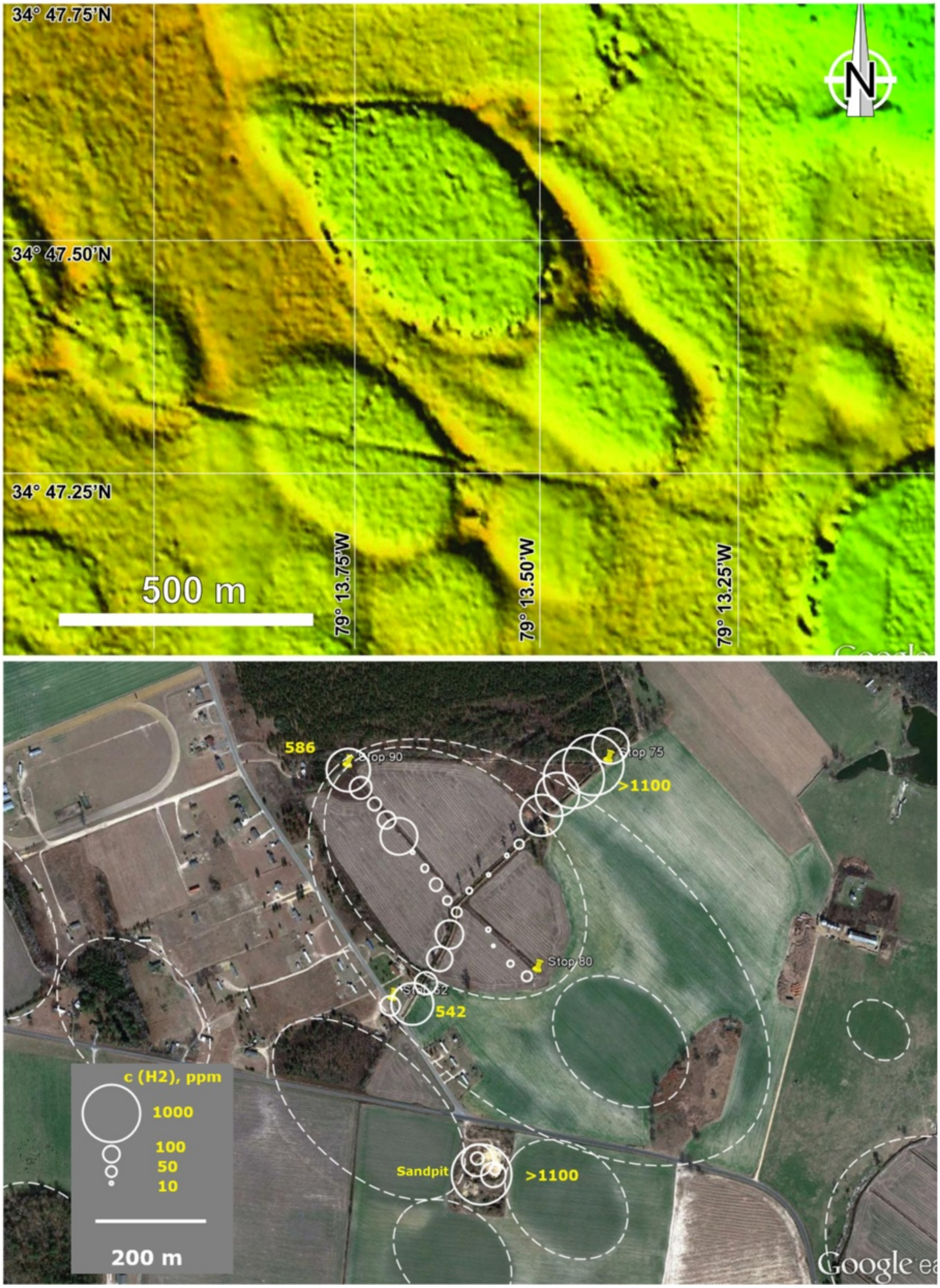

Fig. 2 Subsoil H_2 concentration measurements at Arthur Road Bay and Arthur Road Sandpit (*lower image*). *Dashed lines* outline the bays. *Upper image* is a LiDAR image showing the relief of the lower image. Profile lines follow ditches (*dark lines* in the lower photograph). Date of measurements 14 March 2012. LiDAR image is from the site cintos.org

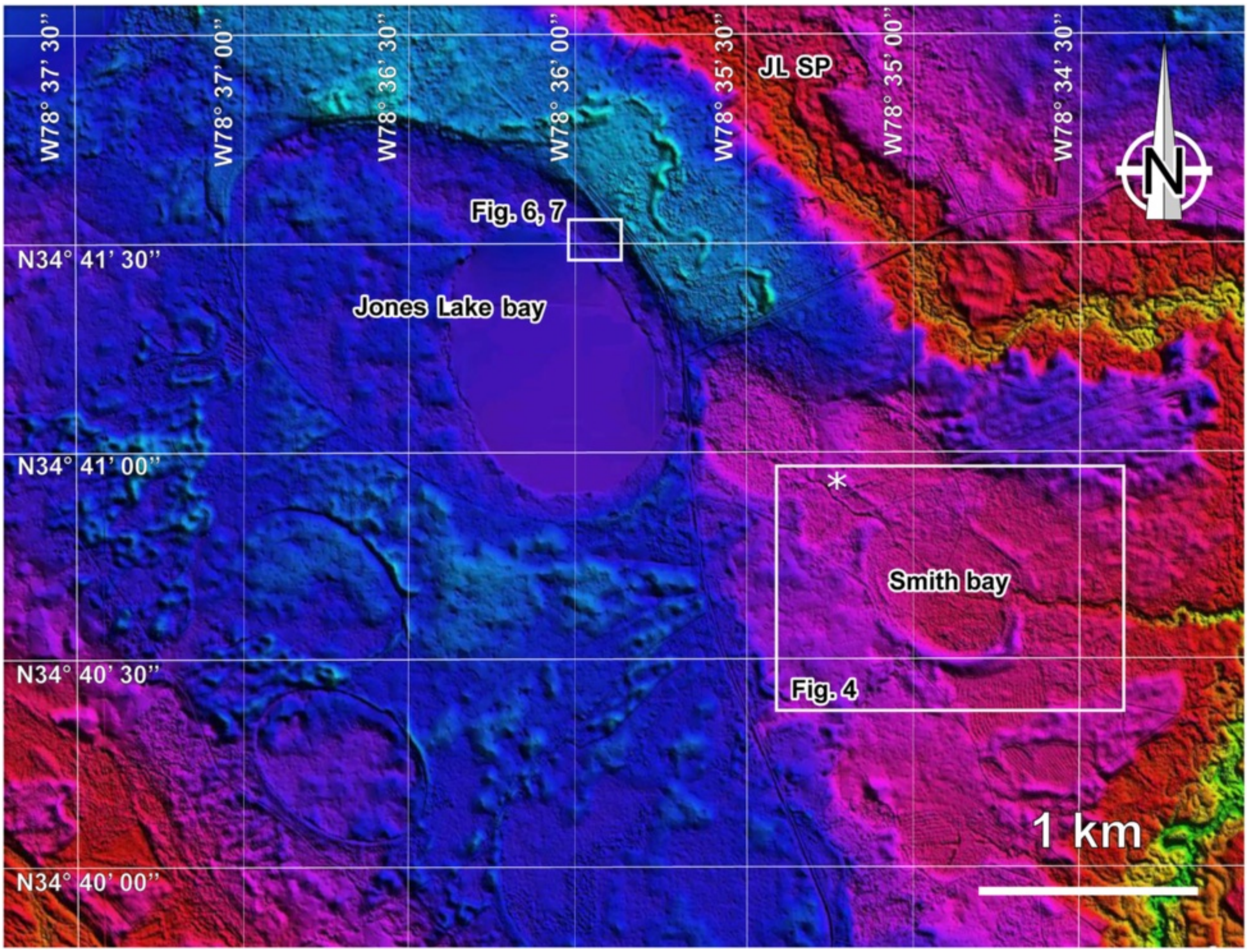

Fig. 3 LiDAR image with the locations of sites shown in Figs. 4, 6, and 7. *JLSP* Jones Lake Sandpit. The *asterisk* is the intersection zone for Smith Bay and unnamed bay. LiDAR image is from the site cintos.org

distribution of the shallow subcrops of the NC Coastal Plain. The features known as Carolina bays postdate the formation of the terraced topography.

Carolina bays are dynamically active features. According to Cohen et al. (1999), they originated during the Pleistocene. A relatively new technique for dating quartz grains, optically stimulated luminescence (OSL), has helped to refine the time frame of bay formation. Ivester et al. (2004) used this dating method to establish the multiple phases of bay evolution during the past 100,000 years. Brooks et al. (2010) concluded that most of the bays formed during the late Pleistocene, and Frey (1954) and Thom (1970) provided evidence for recent enlargement of existing bays. Moore et al. (2014) demonstrated geomorphologically and stratigraphically that these bays migrate through their own lacustrine deposits and calculated the migration rate from a series of OSL dates. In this study, we document an actively forming bay.

Methods

Study sites

The study sites (Figs. 1 and 3 and Table 1) are three prominent Carolina bays (Arthur Road Bay (Fig. 2), Smith Bay (Figs. 3 and 4), and Jones Lake Bay (Fig. 3)), and four smaller-scale detailed study sites associated with them. The detailed study sites include two sandpits (Arthur Road Sandpit and Jones Lake Sandpit), a small, recently formed bay-like feature associated with Jones Lake and a bay intersection area (between Smith Bay and an unnamed bay). All the study sites are on public land in Bladen County, except Arthur Road Bay and the sandpit located in Robeson County, which are privately owned. These public lands include Bladen Lakes State Forest, Turnbull Creek Educational State Forest, and Jones Lake State Park. The bays are naturally well preserved in these areas.

The Arthur Road sites are situated on surficial units that are Pliocene in age. The Smith Bay and Jones Bay

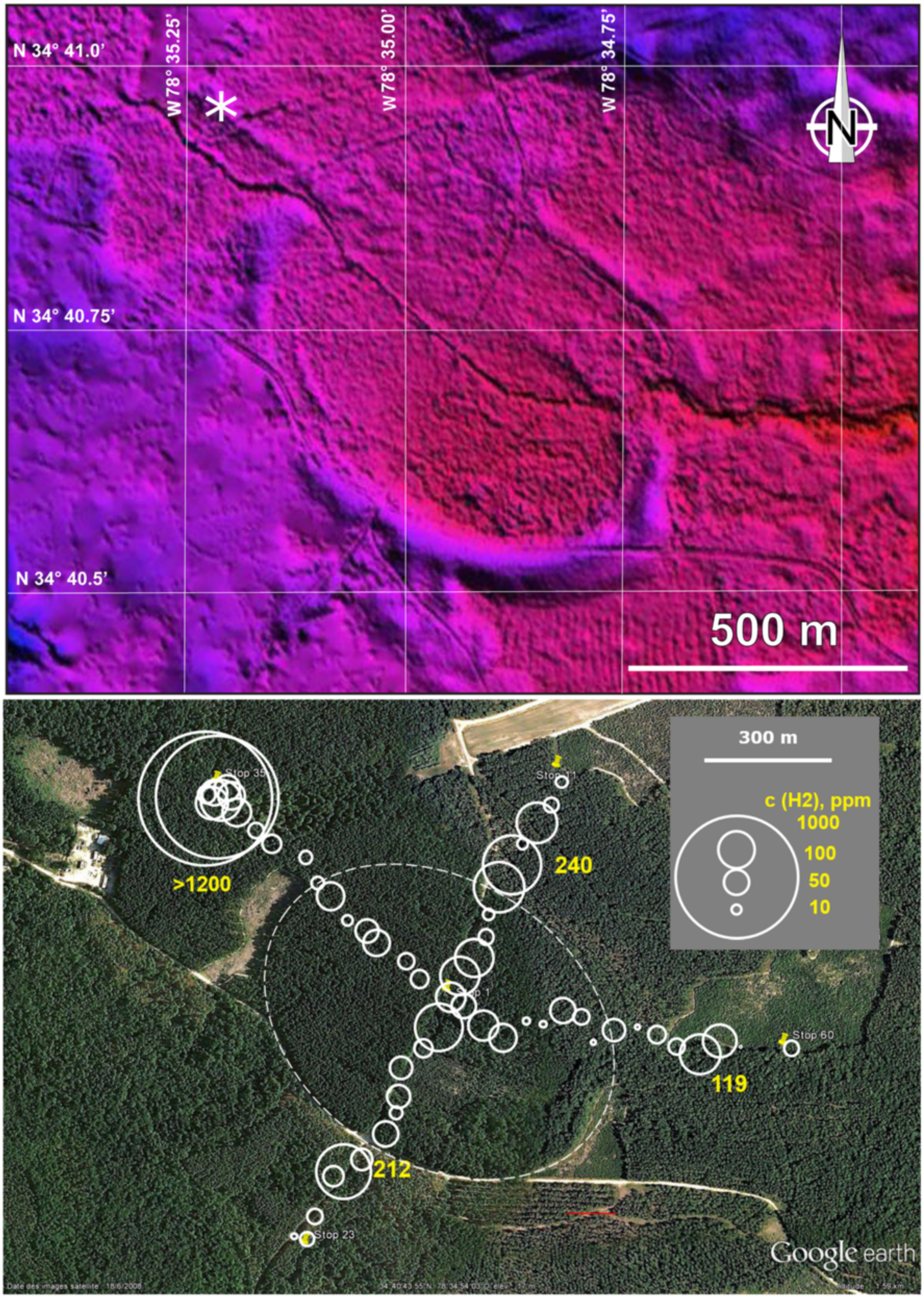

Fig. 4 An example of the subsoil hydrogen concentration measurements made in Smith Bay (*lower image*). *Upper image* is the corresponding LiDAR image. The *asterisk* is the intersection zone for Smith Bay and unnamed bay. *Dashed lines* outline Smith Bay. Date of measurements 09 March 2012. LiDAR image is from the site cintos.org

Table 1 Gas chromatography (GC) measurements of soil gas samples

Work site	Sample index number	°dec N	°dec W	H_2 measured in the field (ppm)	GC measurements						
					H_2 (ppm)	O_2 + Ar (%)	N_2 (%)	CH_4 (ppm)	CO_2 (%)	C_2H_6 (ppm)	C_3H_8 (ppm)
Arthur Road Bay		34.7939167	−79.2296667	586	275	20.05	79.30	11	0.62	1	2
Arthur Road Sandpit		34.7869444	−79.2266667	SAT	605	19.37	79.28	735	1.21	1	1
Arthur Road Sandpit, bubbles		34.7870556	−79.2269167	–	0	2.84	35.04	53.57 %	8.56	0	0
Smith Bay	1	34.6824722	−78.5870694	659	179	20.23	79.26	15	0.50	2	2
	2			715	146	20.32	79.15	11	0.51	0	0
	3			574	296	20.38	79.40	17	0.20	2	2
Small new structure inside Jones Lake Bay	1	34.6930278	−78.6004722	210	107	20.12	79.61	194	0.23	0	0
	2	34.6930556	−78.6008056	391	167	16.51	79.35	27468	1.38	0	0
	3	34.6928131	−78.6003308	477	202	20.00	78.11	5875	1.28	0	0
	4	34.6928232	−78.6003460	815	463	18.67	78.66	13783	1.24	0	0
Jones Lake Sandpit	1	34.7001111	−78.5867222	3700*	698	14.54	84.02	244	1.34	0	0
	2	34.7001111	−78.5872500	719	245	20.34	79.30	15	0.33	0	0
	3	34.7001111	−78.5867222	SAT	1043	14.78	82.12	392	2.96	0	0

Data for all field H_2 measurements were obtained with a GA2000 Plus detector, except in fields marked with *, which were measured with the MDS detector. Total gas measured for each sample is equal to 100 %. Discrepancies between field measurements and laboratory measurements are attributable to the sampling method (see explanation in "Field methods" subsection). Field measurements were usually about twice the laboratory measurements, except where sampling failed. For all field measurements, see Additional file 1: Table SI-1
SAT saturation (overload) of the detector, *°dec* coordinates in decimal degrees

localities occur on Middle to Early Pleistocene deposits (see Abbott et al. 2011 for their Figs. 4 and 5). The surficial units at all the sites overlie Cretaceous formations (see Fig. 6 in Abbott et al. 2011).

Site selection

We chose the study sites using satellite images, by reviewing areas that included a high density of bays with a variety of dimensions, that were easy to access and represented various ages deduced from the geomorphology. We identified the bays using satellite images from Google Earth and Google Earth Pro. LiDAR images were downloaded from the site cintos.org. We measured the H_2 concentrations along profiles that transected the entire bays, starting outside the bay rims at a distance equivalent to half the diameters of the bays. This allowed us to determine the point on the profile at which H_2 started to increase around the bays.

Field methods

Field measurements of the soil gas concentrations were made with a method modified from previously described techniques (Battani et al. 2010; Rogozhin et al. 2010; Garcia et al. 2012; Sukhanova et al. 2013; Larin et al. 2014). Small holes were drilled with a portable perforator using a 120 cm long drilling bit with a 12 mm diameter. A thin steel perforated tube was then inserted into the hole and the upper part was plugged with Plasticine. We adapted the length of the tube (10, 30, 100, or 120 cm) to the depth of the water table. The soil gas samples were collected in Vacutainer® tubes. Before sampling, a vacuum (10^{-4} mbar) was established in each tube using a turbomolecular pump. Experiments with a 2 m long probe were conducted with a Gas Vapor Probe Kit from AMS Inc., American Falls, ID, USA. This kit has a retractable tip, which allows the soil gas to be sampled at a specific depth.

Two hydrogen gas analyzers were used to measure H_2 in the field. The first was a palladium-based metal-insulator semiconductor detector provided by the Moscow Engineering Physics Institute, Russia, which includes three sensors with different ranges of sensitivity to H_2 (0.5–50, 50–1000, and 1000–16,000 ppm). This detector is selective for H_2 and is not affected by H_2S, CO, or any other gas. It has demonstrated excellent efficiency in several studies in which H_2 was measured or monitored (Firstov and Shirokov 2005; Rogozhin et al. 2010; Sukhanova et al. 2013). The second gas analyzer was the landfill GA2000 Plus multi-gas detector by Geotech® Leamington Spa, UK, equipped with a H_2-sensitive electrochemical cell with a sensitivity for H_2 ranging from 1 to 1100 ppm. Water filters (Pall Acro® 50, 0.45 μm) were used to prevent humidity penetrating the detectors. Detector accuracy was first checked in the laboratory using gas mixtures with H_2 concentrations of 100 and 1000 ppm in air-like mixtures (80 % N_2, 20 %

O_2). The relative accuracy of the detectors was "within ±10 %." The results of all field tests are shown in Additional file 1: Table SI-1.

Laboratory analyses

Laboratory analyses were performed to determine the molecular compositions of gas samples in Vacutainer® tubes using gas chromatography (GC), and the results were compared with the measurements made in the field. The chromatograph used was a VARIAN 3800 by Agilent Technologies, Santa Clara, CA, USA, which is equipped with several columns in series and three detectors: two thermal conductivity detectors and one flame ionization detector. The analytical results are given with a precision of ±0.1 %. Each analysis was bracketed with blanks.

The compositions of the collected gas samples showed that air made an important contribution, which is very normal for soil gas. The values determined with the GC analysis were always lower than the field measurements, because of the sampling method used. The samples were measured after the maximum concentration of H_2 was reached on the field detector, when its value started to decrease. Therefore, the samples were always diluted because the soil gas mixes with air present in the H_2 detector system.

Results

Arthur Road Bay

The Arthur Road Bay is an oval-shaped depression (580 m long and 360 m wide) with a sand rim (Fig. 2). It has been modified by anthropogenic activity (draining and clearing). Two ditches drain water from this bay. Data were collected along two profiles that transected the bay perpendicularly (Fig. 2). Gas measurements were made every 50 m along both profiles (Fig. 2). The maximum detected H_2 concentration was >0.11 % (detector saturation), with an average of 233 ppm across 23 measurements (Additional file 1: Table SI-1). Traces of C_2+ hydrocarbons were detected in the laboratory analysis of one soil gas sample (Arthur Road Bay, Table 1).

Arthur Road sandpit

Additional measurements were collected in a sandpit located 500 m south of the Bay's center (Fig. 2). The pit was surrounded to its north, east, and west by at least three other bays (Fig. 2). The maximum H_2 concentration detected at the pit was also >0.11 % (detector saturation), with an average of 313 ppm across six measurements. The laboratory analysis of one soil gas sample provided evidence of C_2+ hydrocarbons (Table 1). Some parts of the sandpit included stagnant water pools that were degassing natural bubbles of gas. A GC analysis revealed a high concentration of methane (54 %) in the bubbles (Table 1), which could suggest high biological activity of methanogens using the H_2.

Smith Bay

Smith Bay is an oval-shaped depression with a sand rim (Figs. 3 and 4). This depression is 1–3 m deep, 720 m long, and 545 m wide. The bay formed on a probable middle Pleistocene landscape. Inside its rim, it is mantled with 1–2 m of Holocene peat. The vegetation patterns, swamp distribution, and the topography sharply delimit the bay's outline and its rim. Water drains from the bay in two ditches that cross near the center of the bay. Data were collected along two profiles that followed the ditches (Fig. 4). A maximum concentration of H_2 >1200 ppm (saturation of the GA2000 detector) was detected outside the northwestern border of the bay (see Fig. 4), near the intersection of Smith Bay and a smaller unnamed bay to the northwest. Three other sites with H_2 peaks >1200 ppm were situated where the transects coincide with the sand rims bordering the bays (Fig. 4). Elevated concentrations of H_2 were also detected in the center of the bay. The H_2 concentration decreased to near-zero values outside the sand rim.

Intersection zone for Smith Bay and unnamed bay

The maximum H_2 concentration (>0.12 %) occurred at a site that coincided with the overlap or intersection of the rims of the two bay (Smith Bay and an unnamed bay situated to the northwest) (Fig. 4). Here, the surface was mantled with peat (70–80 cm thick)-overlying sand. In a small area of the bay overlap where high H_2 concentrations were detected, ten measurements were made within a circumference of 5 m. Only one other point inside this circumference displayed a similarly high H_2 concentration (Additional file 1: Table SI-1). The subsoil H_2 concentrations here were highly variable, probably because the porosity and macropore distribution of the soil were heterogeneous.

We performed an experiment with a 2 m long probe near the recorded maximum H_2 concentration to measure the gradient of the H_2 concentration. Measurements were made at several depths (every 50 cm) using a system of retractable tips (Gas Vapor Probe Kit). An increase in the H_2 concentration with depth (Fig. 5) was observed at the transition from peat to sand (within a 50–100 cm interval), and the maximum concentration was in the coarse sand. As well as H_2 and CH_4 anomalies, C2+ hydrocarbons were also detected in samples from this location (Smith Bay 1–3, Table 1) with laboratory measurements.

Jones Lake Bay

Jones Lake Bay is a very large bay (2400 × 1500 m), with an almost perfect inner elliptical lake (Jones Lake) in the

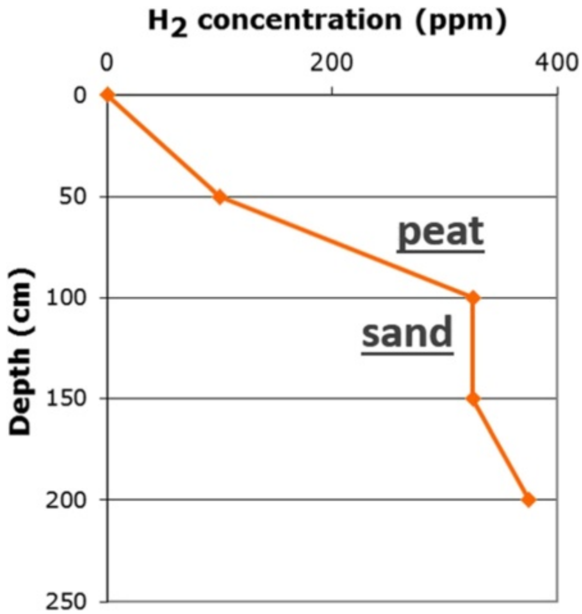

Fig. 5 Increasing hydrogen concentrations with depth in the intersection zone of Smith Bay and unnamed bay. Its location is shown in Fig. 4. Date of measurements 10 March 2012

southeastern part (Fig. 3). Like the other bays studied, the limits of the bay are clearly detectable from the vegetation. The bay is covered with peat and is densely vegetated on the inside, whereas outside it, the soils are sandy and have rare undergrowth. The bay rim consists of soils with coarse sand derived from the underlying deposits of probable Early Pleistocene age. H_2 was only detected within the confines of the bay border. The maximum concentration of H_2 was 815 ppm, detected in coarse sand. We detected no traces of H_2 outside the bay limits (Additional file 1: Figure SI-1 and Additional file 1: Table SI-1).

Jones Lake sandpit

We also measured the H_2 concentrations in a sandpit near the Jones Lake Bay (Fig. 3 and Additional file 1: Figure SI-1, upper right part). We detected very high concentrations of H_2 at the bottom of the peat, 3–4 m below the ground surface, in clayey sand. The maximum concentration was 0.37 % H_2, measured with the Gas Vapor Probe Kit. The average H_2 concentration was 0.1 % across seven measurements (Additional file 1: Table SI-1).

Small new structure inside Jones Lake Bay

Detailed studies were conducted in a small area outside the lake in Jones Lake Bay (Fig. 3, insets in Figs. 6 and 7), where high H_2 levels (up to 815 ppm) were detected in the soil. This site is a relatively small near-circular wet area (diameter ~60 m) where the tall trees suddenly withered naturally in 2008–2009, which was confirmed by the park rangers who manage the reserve. Satellite images taken in 2008 were compared with images taken in 2009 (Fig. 6). The trees probably withered (see panoramic picture, Additional file 1: Figure SI-2) in response to the increase in soil water saturation (water table depth of only 10–15 cm) or from the negative impact of another unknown process affecting the vegetation. Despite the very high water table, an H_2 concentration of 210 ppm was detected in the upper few centimeters (<10 cm) of the soil in the center of the newly forming bay (Fig. 7). The depression of the newly forming bay was less than 1 m, according to field measurements made with an altimeter.

Two detailed cross-sections of the new bay, showing the H_2 concentrations and distances, with measurements made every meter, are shown in Fig. 7. In both sections, the highest H_2 concentrations were detected on the new bay's borders. Laboratory measurements of the chemical composition of the gas revealed that relatively high concentrations of methane were associated with H_2 (Table 1). No heavier hydrocarbons were detected. This wet area with H_2 anomalies is very similar to a recently formed round feature studied by our team in Russia, near the city of Electrostal, close to Moscow (Larin et al. 2014).

Estimation of H_2 fluxes

In this study, the maximum H_2 contents occurred in coarse sand along the rims of the bays and were clearly not associated with wetlands. Because H_2 is a highly diffusive gas, it cannot remain for long in a porous medium-like coarse sand. We used a 2 m long probe (Fig. 5) to demonstrate the gradient in the H_2 concentration, and we interpret this H_2 gradient as the result of the diffusive flow of H_2.

We calculated the H_2 flow using the classical Fick's law of diffusion:

$$F = \Phi \; D \; \text{grad} \; [\text{C}] \tag{1}$$

where F is the gas flow, ϕ is the exchange area or the effective porosity of a homogeneous porous medium, D is the diffusion coefficient of the gas in air, $[C]$ is the gas concentration, and grad is the gradient.

The diffusion coefficient (D) for H_2 in air was taken to be 7.7×10^{-5} m^2/s (Cussler 2009).

The gradients were estimated to be twice the average concentration measured in the perforation, depending on the level of the groundwater. Effective porosity was estimated as follows: for wet swamp, 10–20 %; for dried swamp, 10–25 %; for sand, 35–45 %; and for soil, 25–35 % (Hough 1969; Swiss standard 1999; Das 2008).

To establish the zonation in the H_2 concentrations observed in specific bays, the average H_2 concentrations were estimated for each defined zone in each bay. Four concentric oval-shaped rings were defined for each bay,

Fig. 6 Appearance of new structure inside the Jones Lake Bay. *Upper panel*: satellite image of part of Jones Lake Bay in 2008. *Lower panel*: satellite image of the same part of Jones Lake Bay 1 year later. Its location is shown in Fig. 3. Note the appearance of a *circle* about 60 m in diameter where the forest is damaged

corresponding to the center of the bay, the internal shore, the external shore, and the outside zone. For each zone, the average H_2 concentration was estimated specifically from field measurements. The daily H_2 flow was calculated for each zone, and the sum of the flows calculated for each zone provided the total flow within the bay.

In the Smith Bay, the estimated daily H_2 flow was 750–1000 m^3/day over an area of 1.14 km^2 (660–880 m^3/day/km^2). In the Arthur Road Bay, the estimated daily H_2 flow was 1000–1370 m^3/day over an area of 0.48 km^2 (2240–3060 m^3/day/km^2). For Jones Lake Bay, the daily H_2 flow was estimated to be 1120–2740 m^3/day for a surface area of 6.25 km^2 (180–440 m^3/day/km^2). For the small structure inside the large Jones Lake Bay, the daily H_2 flow was estimated to be 21–31 m^3/day over a surface area of 0.007 km^2 (3000–4400 m^3/day/km^2).

Discussion

The occurrence of molecular hydrogen in the soils of Carolina bays was discovered after noting the remarkable geomorphological similarity to depressions that emit H_2 in the EEC in Russia (Sukhanova et al. 2013; Larin et al. 2014). In both cases, relatively high H_2 concentrations were found in the soil gas of the bay-like features. This study suggests that Carolina bays are geomorphological features related to the occurrence of

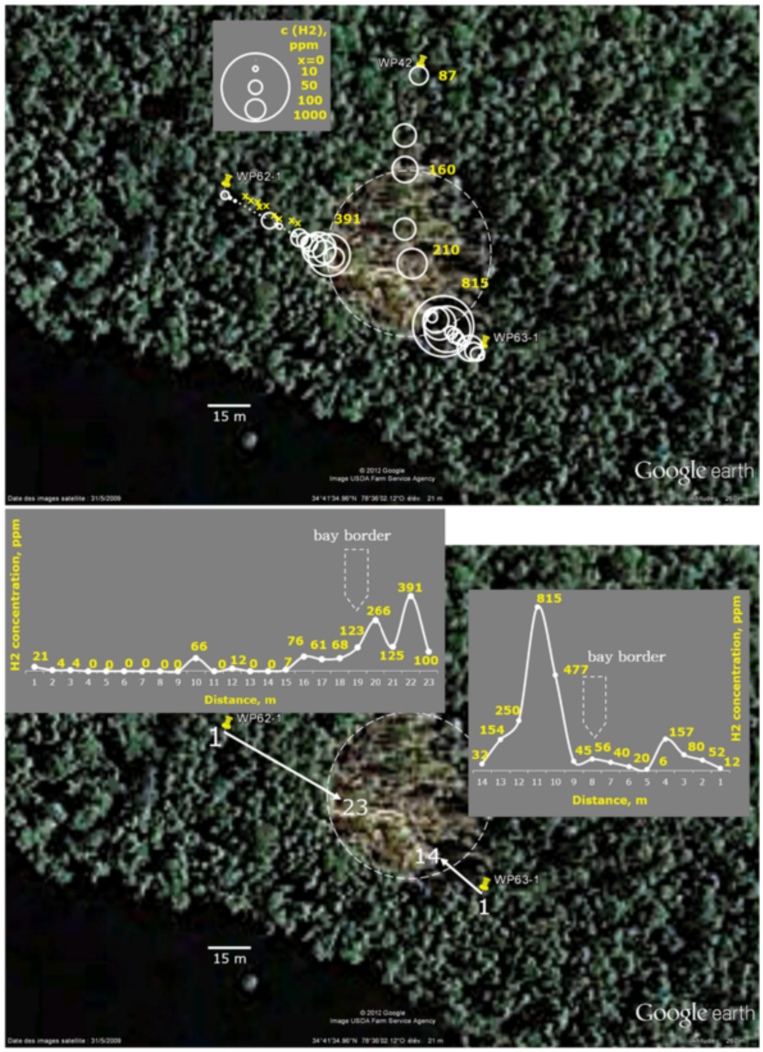

Fig. 7 Subsoil hydrogen concentrations in Jones Lake Bay. Its location is shown in Fig. 3. *Dashed line* shows the limits of the new structure. Geochemistry and geomorphology correlate perfectly, defining the borders of the structure. Date of measurements 13 March 2012

H_2 rather than the results of simple depositional processes along coastal plains (Stolt and Rabenhorst 1987). Therefore, the causal link between the origin of these morphological features and their association with molecular hydrogen must be established. This bay–hydrogen association suggests a connection between fluid seepage from depth towards the atmosphere (deep geological control) and the surficial geomorphic expression of Carolina bays. A possible interpretation might be that Carolina bays are not only focal points for groundwater recharge, as suggested by Grant et al. (1998), but also a morphological expression of fluid seepage from depth toward the atmosphere.

Origin of hydrogen

Carolina bays are natural depressions, commonly filled with water and organic deposits (peat). Consequently, we should consider whether the source of H_2 might be attributable to superficial biological activity under reducing conditions (Sugimoto and Fujita 2006). However, our study clearly shows that the highest concentrations of H_2 systematically occurred in the sand deposits on the rims of the bays, where there was no peat cover (Figs. 2, 4, and 7). We also detected H_2 at a depth of 2 m, 120 cm below the level of the peat (Fig. 5), which reduces the likelihood of a very-near-surface origin for the H_2. Furthermore, the H_2 concentrations measured in the wetlands were low compared with the measurements made in sand. The H_2 concentrations also increased with depth (Fig. 5, Additional file 1: Table SI-1), and the maximum concentration measured, 0.37 vol.%, was detected at a depth of 5 m below ground level in sandpit 2 (Additional file 1: Table SI-1), in a clay and sand substrate. Although slight background H_2 associated with biological activity cannot be excluded, these observations make a purely superficial origin for the H_2 highly improbable. It is also generally believed that the free H_2 produced by bacteria remains at low concentrations because it is rapidly consumed by methanogenic bacteria and soil enzymes (Conrad and Seiler 1981). Indeed, the biological production of H_2 occurs within bacterial consortia, where this valuable chemical energy source is constantly consumed and consequently remains at low concentrations (high H_2 concentrations inhibit H_2-producing bacteria). H_2 is considered to be the most energetic substrate, able to sustain lithoautotrophic ecosystems in subsurface environments, where it is readily consumed (Nealson et al. 2005). Therefore, such high concentrations in soil gases are more probably linked to geochemical H_2-producing processes.

Inventories of the geological controls on the sources of natural hydrogen have been made by Apps and Van De Kamp (1993) and Smith et al. (2005). The natural settings for hydrogen include hydrocarbon-bearing basins, young organic-rich sediments, coal beds, fault zones, extrusive igneous rocks, alkaline igneous complexes, geothermal fields, crystalline basements, potash-bearing strata, salt-bearing strata, and ultramafic rocks. The geologically controlled sources of natural H_2 can be grouped according to four main families of processes: (1) water hydrolysis processes (several processes that include the oxidation of ferrous minerals, radiolysis, cataclasis, and metamorphism); (2) organic matter decay (including thermal maturation); (3) methane and/or ammonia decomposition during metamorphism; and (4) deep Earth degassing. The alteration of Fe(II)-bearing minerals is the most commonly reported source of natural H_2 seepages on Earth, notably at mid-oceanic ridges and in ophiolitic massifs, where mafic and ultramafic are altered. Moreover, a recent study suggested that the H_2 production from the Precambrian continental lithosphere has been underestimated (Sherwood Lollar et al. 2014).

Several ultramafic suites in the eastern Piedmont Province of NC are interpreted as parts of ophiolite sequences (Butler 1989). In particular, the Halifax County complex, described in detail by Kite (1982) and Kite and Stoddard (1984), and interpreted as ophiolitic, is locally overlain by Coastal Plain deposits. Kite and Stoddard (1984) also proposed that an extensive ophiolitic belt might occur beneath the Coastal Plain (also see Lawrence and Hoffman 1993). However, the available deep drilling data are insufficient to define exactly where these rocks are located (and at what depth). The alteration of peridotite, in both oceanic and continental contexts, produces H_2 with the reduction of water by Fe(II), contained especially in fayalite (olivine ferric phase). This process, which is associated with serpentinization, consumes water and is responsible for mineral hydration. Other minerals, as well as olivine, contain Fe(II). In particular, clays (ferrous illites, chlorites, or smectites) can release Fe^{2+} ions in solution under certain conditions. The oxidation of dissolved ferrous ions by water produces H_2, and this type of process could be a potential source of H_2 if the sedimentary pile is clay-rich and provides a substantial reservoir of Fe(II).

It has been suggested that H_2 forms during rock–water reactions (e.g., cataclasis) between fresh rock surfaces containing radicals (Sugisaki et al. 1983) and by the redox conversion of hydroxyls to peroxy groups in silicates (Freund et al. 2002). H_2 can also be formed in uranium-, thorium-, and potassium-rich geological settings, by the radiolysis of water and/or organic matter, and also by the reaction of water with newly formed elements (Savchenko 1958; Lin et al. 2005). However, the quantities of H_2 that can be produced by these mechanisms are limited, and therefore they cannot explain the H_2 flows estimated in this study.

Another possible source of H_2 is the decay of solid or dissolved organic matter, either by bacterial decomposition in sediments during diagenesis and/or later during thermal maturation, which produces H_2 during the ultimate cracking of organic matter. Thermal maturation can be ruled out on the coastal plain of eastern USA because the sedimentary cover of NC and SC is not thick enough to produce the burial depth necessary for H_2 production by thermal maturation.

The decomposition of methane and/or ammonia at high temperatures (above 600 °C) during metamorphism can produce H_2. Such reactions can also produce dinitrogen. In the present case, we observed a diffusive flow of H_2 in the soil, but because it was already mixed with atmospheric components, it was difficult to evaluate the gas components that evolved with it, which may help to clarify the origin of this molecular hydrogen.

The degassing of the Earth's mantle is usually ruled out based on our present understanding of the oxidation state of the upper mantle, and it is therefore not compatible with the high concentrations of dihydrogen detected here. However, because the amount of hydrogen that was originally incorporated into the Earth's interior is unknown, the oxidation state of the upper mantle may actually differ from the conclusions drawn from surface observations, which are necessarily indirect. A recently revised geochemical concept (Toulhoat et al. 2015), initially proposed by Larin (1993), suggests that the Earth's interior is enriched in hydrogen, which is supported by calculations and experiments with iron hydride (Isaev et al. 2007). Such a model is consistent with Ohmoto's suggestion (Ohmoto 1997) that the oxygenation of the atmosphere is linked to the evolution of the continental crust. Under these conditions, deeply stored hydrogen could slowly seep to the surface.

The daily H_2 flows estimated in this study are quite important. For the largest structure, Jones Lake, they varied from 1120 to 2740 m^3/day. If the total number of Carolina bays spread along the Atlantic coast of the USA (about 500,000) is considered, it is clear that a large-scale process is involved, and the mechanism that produces this hydrogen must be efficient enough to sustain the observed H_2 flow.

Geometry and origin of Carolina bay structures

Whatever the source of this hydrogen, the subsurface migration of the ultimate reduced gas must induce reactions with oxidized subsurface rocks and fluids. These reactions and the seepage routes taken might be associated with the initial formation of the elliptical geomorphic structures that are characteristic of Carolina bays. These landscape features develop exclusively in areas where unconsolidated sediments are present at the surface. This is the case in both Carolina and Russia, where the weathered bedrock below the soil consists of unconsolidated granular sediments (Larin et al. 2014). These structures are rarely seen in river valleys, where they are likely to be obscured by fluvial processes. However, a creek that crosses Smith Bay (Fig. 4) does not seem to greatly affect the form of the bay. This may indicate either the recent age of the creek or of Smith Bay.

A variety of compounds can form with the hydrogenation of rocks along the migration pathways of H_2, including water, hydrocarbons, and acids. All these compounds are susceptible to mobilization and to migration out of the reaction zone. In this way, hydrogen might induce an increase in the bulk porosity of the rocks during its vertical migration, and as a consequence, it is likely to create its own channel for vertical migration. All the associated processes (degassing, dewatering, and volume loss at depth) will generate subsidence at the land surface, thus forming rounded (circular or elliptical) depressions.

A study of similar depressions in the Ukraine suggested that water seepage from them is possible (Bixio et al. 2002). Therefore, water with dissolved hydrogen might be discharged onto the surface, creating swamps in some bays, such as the newly formed, small structure in Jones Lake Bay. This small depression, which appeared during the late 2000s, suggests a still active mechanism underlying the formation of some of the bays. Satellite images taken in 2008 show no sign of this structure, whereas 1 year later in 2009, it had become clearly visible (Fig. 6), with the loss of all the trees within it, within less than 1 year, which was probably related to the flooding of the area. The same rapid formation of a new structure, within only several years, has been observed in the Moscow region of Russia (Larin et al. 2014).

The highest concentrations of H_2 occurred on the external slopes of the sandy rims of the Carolina bays, with moderate concentrations inside the bays and no H_2 recorded at some distance from the bay limits (Figs. 2, 4, and 7). This distribution of H_2 is similar to that in the structures studied in the EEC (Larin et al. 2014). The sand rims that border the bays (1–3 m higher than surrounding surface elevation; visible on LiDAR images in Fig. 3 and highlighted in Additional file 1: Figures SI-3 and SI-4) often showed the highest concentrations of H_2. These zones appear to be the preferential drainage sites for H_2-rich fluids moving toward the surface. Although the mechanisms of their formation and their association with higher concentrations of H_2 are not yet understood, they may be linked to fault networks around the features or their peculiar petrological properties (notably the porosity of the rocks surrounding Carolina bays).

Field studies in the EEC have shown that bay-like features emitting H_2 gas sometimes occur along structural trends, very probably corresponding to basement faults (Larin et al. 2014). Many studies have suggested that H_2 anomalies are commonly related to faults (Wakita et al. 1980; Jones and Pirkle 1981; Ware et al. 1985; Sato et al. 1986; Shcherbakov and Kozlova 1986; McCarthy and McGuire 1998; Rogozhin et al. 2010), which act as fluid conduits. This suggests that H_2-emitting features are genetically related to structural features of the crystalline basement. The available information on the deep geology of NC is insufficient to exactly define the locations of faults or their distances from these depressions (Lawrence and Hoffman 1993). This theory, together with the potential alignment of the bays along structural trends, suggests a close relationship between Carolina bays and the observed molecular hydrogen seepages from these potential geological structures.

The elliptical shape of Carolina bays is the feature that most significantly distinguishes them from the hydrogen-seeping structures in the EEC, which usually have rounded shapes. The elliptical shape of Carolina bays could be interpreted as a consequence of the local stress regime. The long and short axes of the bays appear to occur parallel to the minimum and maximum horizontal stresses, as is the case for many calderas. The formation of stress-induced oval structures is well documented in calderas around mud volcano systems (Bonini 2012). Stress discharge will determine the predominant orientation of the vertical (or subvertical) fractures of crystalline basement rocks (Lawrence and Hoffman 1993). Indeed, on geological maps, faults are almost always parallel to the bays' orientation (Brown 1985). Consequently, the initially round (isometric) shape of the hydrogen stream would gradually become elliptical as it ascends through the upper layers of the lithosphere.

When Carolina bays are compared with the structures in the EEC (Larin et al. 2014), the chemical composition of the gases seeping from them and the flow rates of the gases are quite similar. The H_2 concentrations range from tens to hundreds of parts per million. Small quantities of CH_4 and its close homologues (C2+) are also sometimes present locally. We observed similar links between the geochemistry, geomorphology, and the distributions of the H_2 concentrations in the Carolina bays and the Russian structures: the highest concentrations commonly occurred on the external shores of these structures, with high H_2 concentrations inside the structures and almost zero H_2 outside the depressions. According to our estimates, larger bays showed greater absolute H_2 flows, whereas smaller bays showed greater flows per unit of surface.

When soil scientists studied the hydrogen-seeping features in the EEC in Russia (Sukhanova et al. 2013; Polyanskaya et al. 2014), they found that molecular hydrogen seeps from these structures and that this seepage affects the soil layers by disturbing the vegetation and the microbial biomass. In areas of H_2 seepage, the humus content decreases by a factor of 2–3 and the optical density of the humic acids is lower than in the surrounding areas. The fertility of arable lands decreases significantly, and they often become unsuitable for cultivation (Sukhanova et al. 2013). The total quantities and biomasses of bacterial cells and fungal spores and the lengths of fungal and actinomycete mycelia decrease (Polyanskaya et al. 2014). The soil bleaching associated with Carolina bays could also be similar to the bleaching phenomenon associated with the bay-like features in the EEC.

The size distribution of the Carolina bays indicates that the number of bays decreases as the surface area of the bays increases (Semlitsch 2000). In this, their size distribution has the same characteristics as the bay-like features in the EEC (Larin et al. 2014), indicating that they may have a common origin.

In summary, the rounded (elliptical) depressions in the Atlantic Coastal Plain, Province of the USA, and the rounded structures in the EEC show similar geomorphic features and size distributions, and they emit H_2 at similar flow rates. All these similarities suggest a common origin and the same mechanism of formation for these features. We interpret them as the surficial marks of pathways of hydrogen-rich fluid migration. We interpret these bays as the results of local structural collapses associated with the rock alterations induced by H_2-enriched fluid flowing from the crust or deeper. Similar features can be seen in satellite images on all continents (except ice-covered Antarctica), suggesting that other analogous structures exist elsewhere in various settings.

Conclusions

The elliptical depressions known as Carolina bays in the USA are associated with H_2 flows, as observed in similar surface features in the East European craton. This molecular hydrogen probably originates from geochemical processes taking place under the sedimentary pile and migrates towards the surface. If such H_2 migration pathways exist, it is possible that this flow of H_2 induces gas–rock interactions, forming shallow pathways. These pathways might link these H_2 flows with the formation of surficial topographic anomalies that correspond to the Carolina bays, and this process might contribute to the active and ongoing development of these bays today. Our observations provide an alternative explanation to the former controversial theories on the formation of Carolina bays. We estimated the daily hydrogen flow to be quite high, up to 2700 m^3, in some of these features. Evidence of the diffusive flow of H_2 seeping from Carolina bays and its abundance suggests that the role of molecular hydrogen in the processes of the Earth's surface environments must be reconsidered. This reconsideration may influence our understanding of the dynamics and chemical processes of the biosphere and the atmosphere (Syvorotkin 2010).

Additional file

Additional file 1: Supplementary information for article.

Abbreviations

EEC: East European craton; GC: gas chromatography; NC: North Carolina; SC: South Carolina.

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

VZ, VB, ED, and NL proposed the topic and conceived and designed the study. All authors performed the experimental study. VZ, VB, ED, and NL analyzed the data and participated in their interpretation. VB, ED, and KF collaborated with the corresponding author VZ in the preparation of the manuscript. All authors have read and approved the final manuscript.

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