Science Bulletin 70 (2025) 2080–2083

Contents lists available at ScienceDirect

Science Bulletin

journal homepage: www.elsevier.com/locate/scib

Short Communication

Discovery of large-scale natural hydrogen leakage in the Zhangbei Basin, North China

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ARTICLE INFO

Article history:
Received 3 June 2024
Received in revised form 18 October 2024
Accepted 21 October 2024
Available online 30 November 2024

Natural hydrogen, as a naturally generated and sustainable clean energy, has recently attracted considerable attention. In contrast to artificially produced gray, blue, and green hydrogens, natural hydrogen can be continuously extracted from subsurface storages [1]. The deep-seated generation of natural hydrogen represents a potential source of primary energy, assuming that recoverable accumulations in geological formations are sufficiently large [2]. Presently, natural hydrogen has emerged as a focus in the global pursuit of clean energy solutions. Extensive drilling in Mali, the United States, and Australia has resulted in serial exploratory discoveries and independent commercial development of super-enriched hydrogen (H_2, up to 98%) [3,4]. Meanwhile, specific concentrations of H_2 (up to 86%) was obtained from gas boreholes of the Songliao, Chuxiong, Qaidam, Shangdu, and Bohai Bay basins in China [4–6]. Additionally, various H_2 leakage sites were identified through soil gas surveys conducted in sedimentary basins across Brazil, Australia, Mali, the United States, and China [7,8]. Hence, the global discovery of natural hydrogen offers a promising opportunity for resource development. However, specialized prospecting and large-scale discovery of anomalous H_2 leakage have lagged substantially, limiting the understanding of H_2 origin, accumulation and distribution. This study presents the results of a 1:100,000 geochemical survey, revealing significant large-scale H_2 leakage in the Zhangbei Basin of North China. These findings differ from previous documented H_2 leakage sites [8].

The Zhangbei Basin is classified as a Mesozoic-Cenozoic continental rift basin located on the northern margin of North China (Fig. 1a). Subsequent tectonism and magmatism splitted it into two smaller basins. Two sets of buried faults, trending NWW and NE, controlled the formation and evolution of the basin during the late Mesozoic and Cenozoic periods [9]. The early Cretaceous active rift basin is attributed to upwelling of asthenospheric materials along a lithospheric-scale tear fault [10]. In contrast, the Cenozoic basin is influenced by the continuous extension of shallow crust linked to the retreat of the Western Pacific plate in North China. This basin contains early Cretaceous lacustrine-alluvial fan deposits, along with late Oligocene and Neogene meandering river deposits, overflow basalt, fan delta-lacustrine deposits, and alluvial fan deposits. Multi-stage magmatic and volcanic activities have occurred in the basin since the Yanshan Stage. A high concentration of carbon dioxide (CO_2, 97.54%) and a certain amount of H_2 (1.92%) were detected in boreholes from the adjacent Shangdu Basin [5]. In this study, we employed a JH90 portable soil gas detector to measure H_2 (max. 1000 ppm, 1 ppm = 1 cm^3 m^{-3} $=10^{-6}$) and CO_2 (>1000 ppm) through *in-situ* geochemical investigations. Seismic reflection, Bouguer gravity anomaly and remote sensing interpretation were used to characterize the fault structure and stratigraphic features of the basin, as well as to delineate the target area for soil gas measurement. Through iterative experimentation with a self-invented sampling device, we established sampling intervals (<1 km × 0.5 km) and times (∼1 min) for gas data collection (Fig. 1b). A total of 1530 measurements of H_2 and CO_2 were obtained from shallow boreholes at a depth of 1.5 m within 1060 km^2 area surrounding Angulinao in the basin, revealing anomalous H_2 and CO_2 concentrations (Fig. 1b, c).

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https://doi.org/10.1016/j.scib.2024.11.025

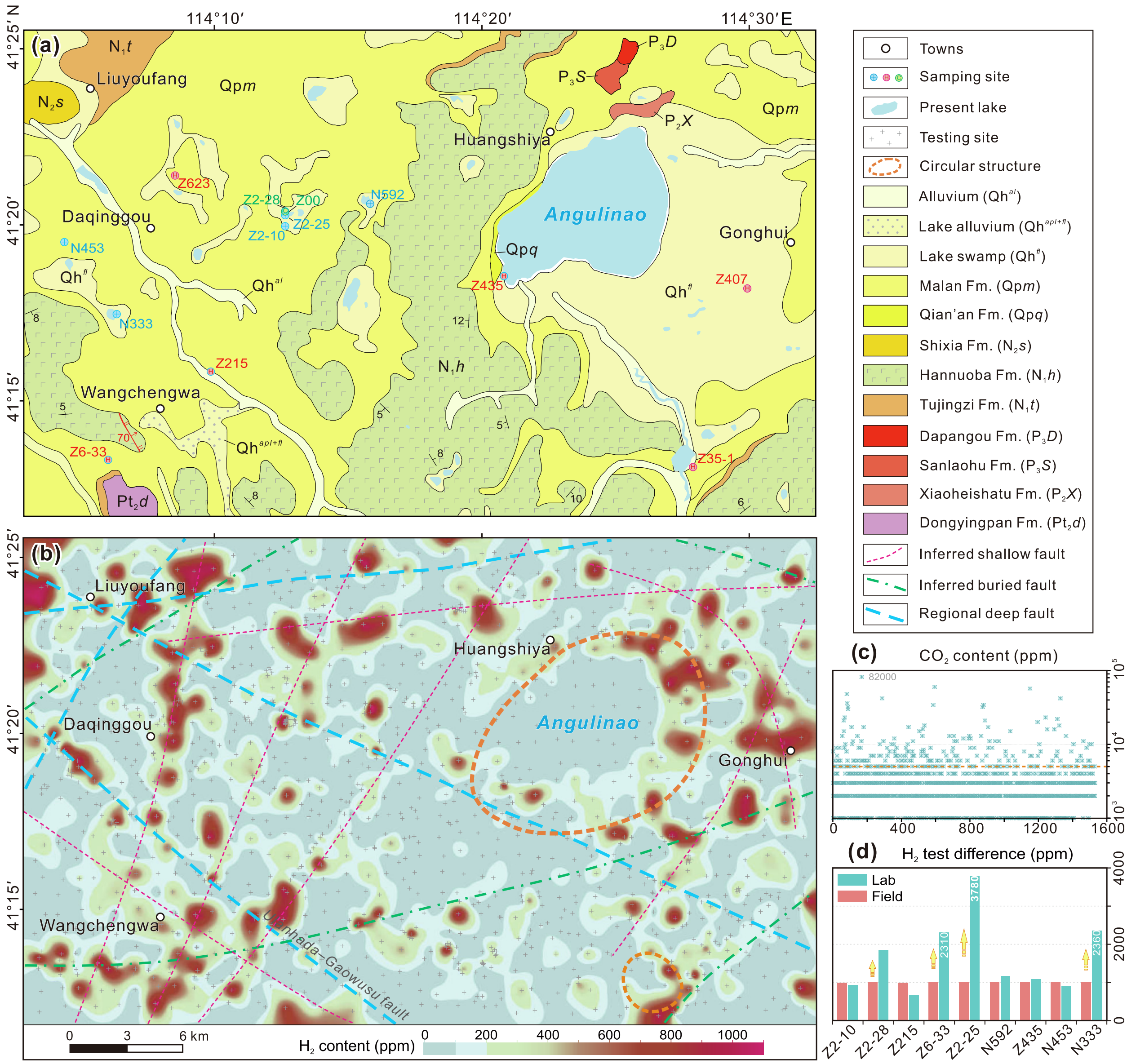

Fig. 1. Investigation results of natural hydrogen in the Zhangbei Basin, including 1:250,000 geological map (a), contour map of hydrogen concentration (b), carbon dioxide concentration (c), and error analysis from field and experimental tests (d).

Our results exhibit elevated levels of H_2, including 76 readings exceeding 1000 ppm, 107 readings falling between 500 ppm and 1000 ppm, and 260 readings ranging from 200 ppm to 500 ppm (Fig. 1b). The contour map illustrates an anomalous H_2 distribution around the intersection of various faults and circular structures (Fig. 1b). Meanwhile, higher concentrations of CO_2, with a maximum of 82,000 ppm and 272 readings exceeding 5000 ppm, were detected in soil gas (Fig. 1c). Furthermore, gas chromatographic analysis of 9 gas samples (GB/T 13610-2014) revealed dominant levels of nitrogen (~79.1%) and oxygen (~20.5%), followed by CO_2 and H_2. Experimental tests of H_2 contents generally align with field results (Fig. 1d); however, they occasionally exceed field limits, reaching a maximum value of 3780 ppm (Fig. 1d). This finding suggests the presence of massive H_2 leakage in the basin. Thus, *in-situ* geochemical measurements of soil gas indicated that the Zhangbei Basin has higher concentrations of H_2 leakage than those found in Brazil (541 ppm), Australia (96 ppm), the United States (815 ppm), Mali (600 ppm) and Borisoglebsk of Russia (3000 ppm). However, these levels are lower than those observed in Moscow of Russia (8000 ppm), and the Sanshui Basin in China (6948 ppm) (Fig. 2a) [7,8]. Notably, an exceptionally high H_2 content (38.4%) was detected in the soil gas of the Amur Basin in Turkey, situated within an ophiolite background (Fig. 2a) [7]. Actually, elevated levels of H_2 are commonly discovered on the edge of plate subduction or collision zones and its peripheral basins, as well as rift basins associated with continental rift systems [11]. These findings highlight the significant potential of natural hydrogen globally.

Identifying the source of H_2 is crucial for research on hydrogen accumulation. The H–C isotope compositions of soil gas samples were analyzed in the laboratory. Our results show that the hydrogen isotope (δD) values of separated H_2 vary between −462‰ and −568‰ (avg. −504‰) (Fig. 2b). The carbon isotope values of abundant CO_2 ($\delta^{13}C_{CO2}$) and trace CH_4 ($\delta^{13}C_{CH4}$) range from −14.6‰ to −22.5‰ and −51.7‰ to −59.2‰, respectively. In this study, we hypothesized that anomalous H_2 concentrations in soil gases originate from deep levels. Firstly, unusually high H_2 contents (Fig. 2a) exceed the background soil H_2 value (<5 ppm) [8]. This exceed

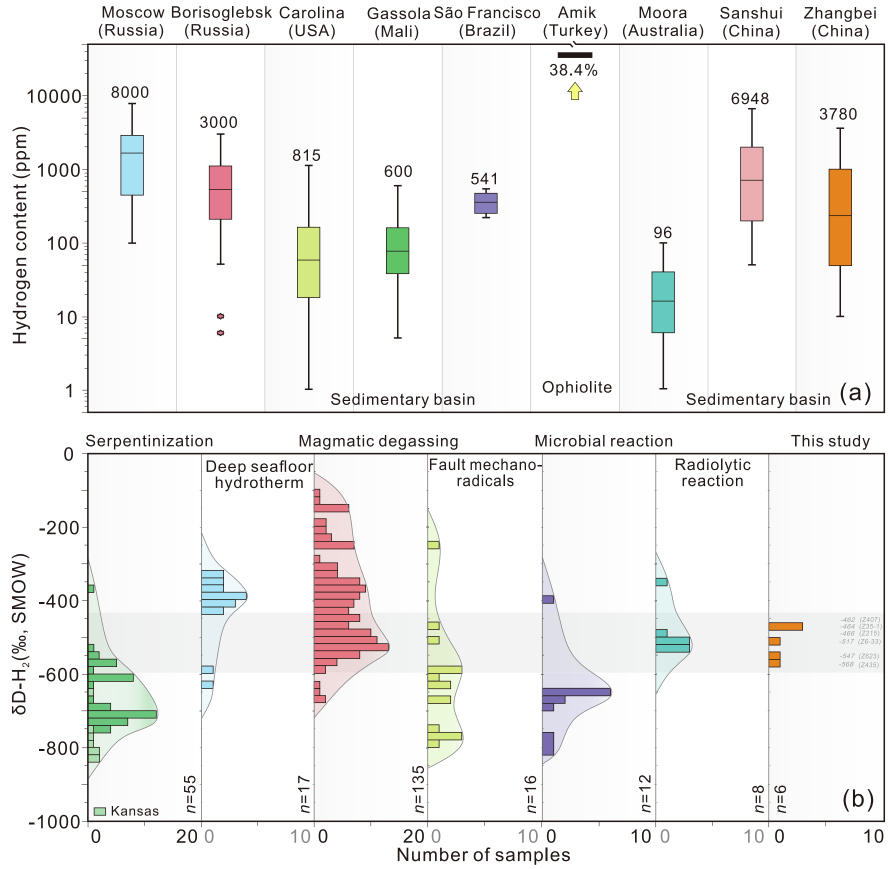

Fig. 2. Discovery of natural hydrogen worldwide, showing hydrogen contents of soil gases at different seepage sites (modified after Refs. [7,8]) (a), and δD values of hydrogen from various sources (modified after Refs. [8,16]) (b).

excludes surface processes associated with microbial activity and plant cover. Secondly, super-enriched H_2 concentrations are accompanied by CO_2 anomalies (max. 82,000 ppm) in the soil gas of study area, which greatly exceed typical soil CO_2 contents. The identifying super-enriched CO_2 reservoir from boreholes (97.54%) in the Shangdu Basin indicates the presence of a deep gas source [9]. Finally, elevated H_2 contents are observed at the intersection of various faults and circular structures, forming NE- or SN-trending bands (Fig. 1b). This implies that deep active faults may influence H_2 migration and leakage. Notably, frequent great earthquakes ($Ms \geq 6$) occurred in the Zhangbei area, resulting in substantial releases of stored H_2 (10,440 ppm) and He (10,370 ppm) [12]. This indicates that the activity of deep faults triggers the release of deep-sourced gases.

The high levels of deep-sourced H_2 are primarily generated by the serpentinization of iron-rich rocks, followed by mantle degassing and water radiolysis [1,4]. Our analysis of regional geology and isotope geochemistry suggests the potential sources of soil H_2 are predominant magmatic degassing and water–rock interactions, and limited water radiolysis in the study area (Fig. 2b). Firstly, multi-cycle tholeiite and alkaline basalts were widely distributed in the Zhangbei area around 33, 22.8–22.1, and 12.2–9.4 Ma (Fig. 1a) [13]. The increased Yb values and reduced La/Yb ratios suggest Cenozoic lithosphere thinning in North China [13], inducing frequent magmatic and volcanic activities. This process may contribute to H_2 production through magmatic degassing from the mantle. A certain amount of H_2 (5.7%–10.9%) was detected in Hannobar basalt inclusions [5], suggesting a probable H_2 source in the deep Earth. Then intense volcanic activities associated with large-scale Hannobar basalt resulted in the release of deep volatiles (e.g., H_2 and CO_2; Fig. 1c, d) during magma cooling and depressurization [8]. Our δD values (−462‰ to −568‰) are consistent with data from regions experiencing mantle magmatic degassing (Fig. 2b). Other isotope data, such as $\delta^{13}C_{CO2}$ (−5.8‰ to −6.5‰), $^3He/^4He$ (3.23×10^{-6} to 3.35×10^{-6}), and R/Ra (2.31 to 2.39) from boreholes of the adjacent Shangdu, also suggest a preferred mantle source [5]. Secondly, we identified a series of Archean pyroxene peridotite and Cenozoic alkaline basalt in the field outcrops. The widespread development of basic to ultramafic rocks may facilitate the production of H_2 during serpentinization. Alternatively, aeromagnetic anomalies in the study area indicate the existence of Precambrian iron-rich basement rocks (e.g., biotite gneiss and iron formations) in the North China [14]. The hydration of iron-rich rocks, coupled to H_2O reduction, may also be responsible for H_2 generation [15], as observed in Kansas [16]. Moreover, shallowly buried Archean iron-rich basement rocks (max. 1800 m) in the Zhangbei Basin are susceptible to water–rock interaction, leading to an increase in H_2 content [4]. Finally, our δD values overlap with data for regions of water radiolysis (Fig. 2b) [16]. The presence of numerous Permian-Jurassic granites in the study area (Fig. 1a) suggests that water radiolysis may serve as a potential source of H_2. Therefore, further studies are necessary to elucidate the H_2 origin, considering the isotopic uncertainty and geological complexity [8].

This study suggests that various active faults and extensive overflow basalt overflow conditions for the migration and accumulation of H_2. The NWW-trending deep active faults, such as Ulanhada-Gaowusu fault, characterized by multi-stage magma distribution during the late Luliang period (Fig. 1b). In contrast, the NE-trending secondary faults coinciding with Hannobar basalt developed during the Himalayan period (Fig. 1b). The deep active faults that intersect the Archean strata suggest their extension into the basement of the late Mesozoic basin [5]. This promotes the long-distance migration of deep-seated H_2 and anomalous leakage along deep faults, followed by dispersion into the basin reservoir via secondary faults (Fig. 1b). Moreover, frequent earthquakes have occurred in the Zhangbei area since the Oligocene, which caused various faults to unseal and release high levels of H_2 [16]. Thus, deep active faults enable the migration of deep-seated H_2 to the basin, while secondary faults then transport H_2 to appropriate reservoirs. Furthermore, low contents of H_2 were observed in exposed basalt areas (Fig. 1a, b), indicating a possible capping effect by multi-stage tight basalts. Additionally, lacustrine and deltaic sandstones from the early Cretaceous and late Cenozoic periods possess the potential to store H_2 via secondary transport along shallow faults. Layered structures with sandstone-mudstone-coal or sandstone-basalt assemblages may serve as effective reservoir-cap combination for H_2 storage, similar to those found in Mali (e.g., sandstonediabase-carbonate assemblage) [3]. Thereby, we assume that tight basalts can be utilized as high-quality cap rock for H_2 storage. Although free hydrogen exists widely in sedimentary reservoirs, dissolved hydrogen in water-bearing reservoirs should require sufficient attention (e.g., Mali) [3]. Generally, the solubility of H_2 in pure water significantly increases with pressure [17], and excess H_2 may form an underwater gas reservoir upon saturation. Consequently, dissolved hydrogen may potentially exist in confined water reservoirs.

In summary, *in-situ* geochemical investigation confirmed anomalous leakage and spatial distribution of large-scale natural hydrogen in the Zhangbei Basin. Elevated levels of H_2 seepage may suggest the presence of vast accumulation or a constant supply of deep-seated H_2. Anomalous H_2 leakage is reasoned to originate from primary magmatic degassing and serpentinization, with limited water radiolysis. Additionally, deep active faults and extensive multi-stage basalts contribute to the migration and distribution of H_2. Dissolved hydrogen may potentially occur in confined water reservoirs. Nonetheless, the formation and identification of H_2 reservoir remains a challenge. Therefore, specialized exploration and research into the extensive discovery and accumulation of natural hydrogen remain nascent, particularly in assessing its potential for commercial development. This progress in identifying large-scale H_2 leakage and distribution underscores the importance of natural hydrogen development in China.

Conflict of interest

The authors declare that they have no conflict of interest.

Acknowledgments

This work was supported by the National Natural Science Foundation of China (42372138), the Basic Scientific Research Fund of the Ministry of Science and Technology of China (J2304), and the Project of China Geological Survey (DD20221807). We thank the Xisheng (Beijing) Energy Co., Ltd, the Beijing Geological Exploration and Development Group Co., Ltd, and the Third Geological Brigade of Hebei Bureau of Geology and Mineral Resources for their assistance during field investigation and experimental work. We gratefully acknowledge *Science Bulletin* Associate Editor-In-Chief, detailed comments and constructive criticism from four anonymous reviewers that greatly improved this manuscript.

Author contributions

Jianliang Jia, Minjie Lu, and Huiqun Yang conceptualized the study. Bingchuan Yin and Zhuangwei Luo performed the methodology. Jianliang Jia, Minjie Lu, Ling Liu, and Qianning Tian performed the investigation and visualization. Yanming Wan, Gang Fu, Yanming Liu, and Huiqun Yang were responsible for supervision. Jianliang Jia wrote the original draft. Jianliang Jia, Minjie Lu, Qianning Tian, Bingchuan Yin, and Zhuangwei Luo carried out the review and editing.

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