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Global aquifers dominated by fossil groundwaters but wells vulnerable to modern contamination

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The Supplementary Information is divided into four sections: S1) compiling global groundwater isotope data, S2) outcomes of young and old groundwater calculations, S3) known biases in our fossil groundwater calculations, and S4) groundwater well construction records in the western USA.

S1. Compiling global groundwater isotope data

Global groundwater isotope data was compiled from hundreds of primary literature sources and from the United States Geological Survey's Water Quality Portal (www.waterqualitydata.us). The raw data sources are shown in Tables S1 and S2. Compiled groundwater carbon isotope data and their associated aquifer latitude and longitude are tabulated and provided as a supplementary dataset (locations are presented to the nearest degree because some sampling locations are known only to ±10s of kilometres).

Groundwater aquifer systems were delineated using geospatial data from the World-wide Hydrogeological Mapping and Assessment Programme (WHYMAP: www.whymap.org). However, the aquifers delineated by WHYMAP were much larger than the areas explored by individual studies in most cases. Therefore, we delineated the aquifer areas surrounding the groundwater isotope measurement locations reported in individual studies, using WHYMAP aquifer boundaries for guidance where possible.

S2. Outcomes of young and old groundwater calculations

The set of Figures S7 (n=62) present the raw, compiled radiochemical (^{3}H and ^{14}C activities) and stable isotope (^{13}C/^{12}C and ^{18}O/^{16}O ratios) data for the study aquifers, as well as the calculated post-1953 and Holocene groundwater fractions. The Holocene groundwater fractions are related to fossil groundwater by [Holocene groundwater fraction] = 1 – [Fossil groundwater fraction].

The uppermost three panels show profiles of geochemical parameters: tritium activities (^{3}H; upper left), radiocarbon activities (^{14}C of dissolved inorganic carbon; upper middle) and stable carbon isotope compositions (δ^{13}C of dissolved inorganic carbon; upper right). The carbon isotope data are for inorganic carbon species dissolved in the water. The lower three panels present the calculated post-1953 groundwater fractions (lower left), Holocene groundwater fractions (lower middle), and stable oxygen isotope compositions (δ^{18}O; lower right). The stable isotope data are presented in delta notation, where $\delta^{18}O = (^{18}O/^{16}O_{\text{sample}}) / (^{18}O/^{16}O_{\text{standard ocean water}} - 1)\times 1000$, and $\delta^{13}C = (^{13}C/^{12}C_{\text{sample}}) / (^{13}C/^{12}C_{\text{Pee Dee Belemnite}} - 1)\times 1000$.

The two panels presenting post-1953 and Holocene groundwater fractions (lower left and lower middle panels) show multiple points per water sample to convey calculation uncertainties. The large symbols show the midpoint estimate for the calculated post-1953 and Holocene groundwater fraction. Smaller-sized symbols represent the upper and lower (i.e., estimated minimum and estimated maximum) fractions of post-1953 and Holocene groundwater.

For all panels, squares represent the midpoint of the screened (i.e., perforated) interval (or top of screen if the midpoint was not given), whereas diamonds represent the total depth of the well. If information for both the top and the bottom of the well was available, we plotted the point twice, once showing the midpoint depth (squares) and once showing the total depth (diamonds) in order to convey the impact of using total well depths rather than screen midpoint depths for our calculations. Red symbols mark isotope measurements of dissolved inorganic carbon.

S3. Known biases in our fossil groundwater calculations

Our findings are likely to be conservative estimates of fossil groundwater percentage and young-old mixing. Our results may be affected by a number of biases, the majority of which would tend to bias our results toward lower fossil groundwater fractions; therefore, our first main conclusion—that fossil groundwater dominates global groundwater storage—should likely be at least as strong as it appears to be.

First, our groundwater ^{14}C dataset is necessarily weighted toward permeable sedimentary basins because these tend to have greater groundwater resources and thus more groundwater wells. These permeable basins plausibly support deeper penetration of younger, Holocene-aged waters than less geologically permeable regions where groundwater drilling is rarer and thus groundwater ^{14}C data are less available. Thus, the radiocarbon data used in this analysis may be biased towards areas with deeper young groundwater, biasing our calculations toward small fossil groundwater abundances.

Second, recent work has pointed out that groundwater samples exposed to the atmosphere before being measured are vulnerable to contamination by modern atmospheric $^{14}CO_2$, which would bias any such contaminated samples in the direction of smaller apparent fossil groundwater fractions[26].

Third, widespread irrigation-induced recharge has likely drawn shallower and generally younger groundwaters deeper, plausibly causing our analysis to underestimate fossil groundwater abundance because our dataset is biased toward irrigated regions.

Fourth, groundwater pumping preferentially draws water from more productive geologic strata, which are more likely to host relatively fast-flowing (and consequently younger) groundwaters, again biasing our analysis towards smaller fossil groundwater fractions.

Fifth, it is possible that well pumping could cause upward flows of fossil groundwaters. Among the aforementioned biases, this case is perhaps the most likely to lead us to overestimate fossil groundwater storage because the stored fossil groundwater has been drawn anomalously shallow close to the well.

Most of the potential biases in our analysis lead us to underestimate fossil groundwater storage. Therefore, our estimate that fossil groundwater comprises most (42-85%) global groundwater storage is likely to be an underestimate.

S4. Groundwater well construction records in the western USA

We compile and present well construction depth data compiled from five western states: (i) California, (ii) Colorado, (iii) Kansas, (iv) Nebraska, and (v) Wyoming.

(i) California groundwater well completion report data were obtained by personal communication with a representative from the California Department of Water Resources (www.water.ca.gov; data obtained during March and April, 2016). The California groundwater well construction records are incomplete in the San Joaquin Valley because data for some counties remains unavailable.

(ii) Colorado groundwater well applications data were accessed from cdss.state.co.us/GIS/Pages/AllGISData.aspx (accessed February 21, 2016).

(iii) Kansas water well completion data were obtained by personal communication with a representative from the Kansas Geological Survey (www.kgs.ku.edu; data obtained during March and April, 2016).

(iv) Nebraska registered groundwater well data were accessed from www.ose.state.nm.us/GIS/geospatial_data.php (accessed February 21, 2016).

(v) Wyoming groundwater well construction data were accessed from sites.google.com/a/wyo.gov/seo/documents-data/maps-and-spatial-data (accessed February 21, 2016).

We focus our analysis on wells designed to extract groundwater for use by households, municipalities, industries and food producers. The compiled groundwater well databases include the stated purposes for which the wells were constructed. We retained wells with purposes that suggest the wells were drilled for industrial, household, municipal and agricultural use (Table S3). We removed wells that we could not confirm to have been installed for industrial, household, municipal or agricultural use.

Table S1. Groundwater isotope data sources

Country	Aquifer	Lon.	Lat.	* Fossil groundwater depth (m)		Primary source of radiocarbon and tritium data
Australia	Ngalia-Amadeus	132	−23	15	70	Cresswell, R., Wischusen, J., Jacobsen, G., Fifield, K. Assessment of recharge to groundwater systems in the arid southwestern part of Northern Territory, Australia, using chlorine-36. Hydrogeology Journal, 7, 393-404 (1999). Wischusen, J. D. H., Fifield, L. K., Cresswell, R. G. Hydrogeology of Palm Valley, central Australia; a Pleistocene flora refuge? Journal of Hydrology, 293, 20-46 (2004).
Australia	Western Port	145	−38	30	75	Currell, M., Cendon, D. I., Cheng, X. Analysis of environmental isotopes in groundwater to understand the response of a vulnerable coastal aquifer to pumping: Western Port Basin, south-eastern Australia. Hydrogeology Journal, 21, 1413-1427 (2013).
Bangladesh	Bengal Basin	91	23	220	250	Aggarwal, P. K., Froehlich, K., Basu, A. R., Poreda, R. J., Kulkarni, K. M., Tarafdar, S. A., Ali, M., Ahmed, N., Hussain, A., Rahman, M., Ahmed, S. R. Isotope hydrology of groundwater in Bangladesh: implications for characterization and mitigation of arsenic in groundwater. IAEA-TC Project Report: BGD/8/016, International Atomic Energy Agency, Vienna, 65 pp. (2000). Hoque, M. A., Burgess, W. G. 14C dating of deep groundwater in the Bengal Aquifer System, Bangladesh: implications for aquifer anisotropy, recharge sources and sustainability. Journal of Hydrology, 444, 209-220 (2012). Klump, S., Kipfer, R., Cirpka, O. A., Harvey, C. F., Brennwald, M. S., Ashfaque, K. N., Badruzzaman, A. B. M., Hug, S. J., Imboden, D. M. Groundwater dynamics and arsenic mobilization in Bangladesh assessed using noble gases and tritium. Environmental Science and Technology, 40, 243-250 (2006). Sikdar, P. K., Sahu, P. Understanding wetland sub-surface hydrology using geologic and isotopic signatures. Hydrology and Earth System Sciences, 13, 1313-1323 (2009). McArthur, J. M., Banerjee, D. M., Sengupta, S., Ravenscroft, P., Klump, S., Sarkar, A., Dischm B., Kipfer, R. Migration of As, and ^{3}H/^{3}He ages, in groundwater from West Bengal: Implications for monitoring. Water Research, 44, 4171-4185 (2010). Majumder, R. K., Halim, M. A., Saha, B. B., Ikawa, R., Nakamura, T., Kagabu, M., Shimada, J. Groundwater flow system in Bengal Delta, Bangladesh revealed by environmental isotopes. Environmental Earth Sciences, 64, 1343-1352 (2011). Dowling, C. B., Poreda, R. J., Basu, A. R., Peters, S. L., Aggarwal, P. K. Geochemical study of arsenic release mechanisms in the Bengal Basin groundwater. Water Resources Research, 38, 1173 (2002).
Botswana	Kalahari Desert	25	−22	150	200	Stadler, S., Osenbrück, K., Suckow, A. O., Himmelsbach, T., Hötzl, H. Groundwater flow regime, recharge and regional-scale solute transport in the semi-arid Kalahari of Botswana derived from isotope hydrology and hydrochemistry. Journal of Hydrology, 388, 291-303 (2010). Mazor, E., Verhagen, B. T., Sellschop, J. P. F, Robins, N. S., Hutton, L. G. Kalahari groundwaters: their hydrogen, carbon and oxygen isotopes. International Atomic Energy Proceedings, Vol. 1, IAEA/CM/182/18, 202-223 (1974). Rahube, T. B. Recharge and groundwater resources evaluation of the Lokalane-Ncojane basin (Botswana) using numerical modelling. Thesis,

Country	Aquifer	Lon.	Lat.	* Fossil groundwater depth (m)		Primary source of radiocarbon and tritium data
						International Institute for Geo-information Science and Earth Observation, 119 pp. (2003).
China	Hexi Corridor (East)	102	39	100	250	Gates, J. B., Edmunds, W. M., Darling, W. G., Ma, J., Pang, Z., Young, A. A. Conceptual model of recharge to southeastern Badain Jaran Desert groundwater and lakes from environmental tracers. Applied Geochemistry, 23, 3519-3534 (2008). Zhu, G. F., Su, Y. H., Feng, Q. The hydrochemical characteristics and evolution of groundwater and surface water in the Heihe River Basin, northwest China. Hydrogeology Journal, 16, 167-182 (2007). Edmunds, W. M., Ma, J., Aeschbach-Hertig, W., Kipfer, R., Darbyshire, D. P. F. Groundwater recharge history and hydrogeochemical evolution in the Minqin Basin, North West China. Applied Geochemistry, 21, 2148-2170 (2006). Ma, J., Pan, F., Chen, L., Edmunds, W. M., Ding, Z., He, J., Zhou, K. Huang, T. Isotopic and geochemical evidence of recharge sources and water quality in the Quaternary aquifer beneath Jinchang city, NW China. Applied Geochemistry, 25, 996-1007 (2010).
China	Laizhou Bay	119	37	70	175	Han, D., Kohfahl, C., Song, X., Xiao, G., Yang, J. Geochemical and isotopic evidence for palaeo-seawater intrusion into the south coast aquifer of Laizhou Bay, China. Applied Geochemistry, 26, 863-883 (2011).
China	North China Plain	115	38	200	300	Chen Z., Nie, Z., Zhang Z., Qi J., Nan Y. Isotopes and sustainability of ground water resources, North China Plain. Groundwater, 43, 485-493 (2005). Kreuzer, A. M., von Rohden, C., Friedrich, R., Chen, Z., Shi, J., Hajdas, I., Kipfer, R., Aeschbach-Hertig, W. A record of temperature and monsoon intensity over the past 40 kyr from groundwater in the North China Plain. Chemical Geology, 259, 168-180 (2009). von Rohden, C., Kreuzer, A., Chen, Z., Kipfer, R., & Aeschbach-Hertig, W. Characterizing the recharge regime of the strongly exploited aquifers of the North China Plain by environmental tracers. Water Resources Research, 46, W05511 (2010). Chen, Z., Jixiang, Q., Jianming, X., Jiaming, X., Hao, Y., Yunju, N. Paleoclimatic interpretation of the past 30 ka from isotopic studies of the deep confined aquifer of the North China plain. Applied Geochemistry, 18, 997-1009 (2003).
China	Songnen Plain	125	49	125	210	Chen, Z., Wei, W., Liu, J., Wang, Y., Chen, J. Identifying the recharge sources and age of groundwater in the Songnen Plain (Northeast China) using environmental isotopes. Hydrogeology Journal, 19, 163-176 (2011).
Germany	Benkerstein	11	49	0	120	van Geldern, R., Baier, A., Subert, H. L., Kowol, S., Balk, L., Barth, J. A. C. Pleistocene paleo-groundwater as a pristine fresh water resource in southern Germany – evidence from stable and radiogenic isotopes. Science of the Total Environment, 496, 107-115 (2014).
Denmark	Ribe Formation	9	56	190	290	Hinsby, K., Harrar, W. G., Nyegaard, P., Konradi, P. B., Rasmussen, E. S., Bidstrup, T., Gregersen, U., Boaretto, E. The Ribe Formation in western Denmark - Holocene and Pleistocene groundwaters in a coastal Miocene sand aquifer. Geological Society of London, Special Publications, 189, 29-48 (2001).
Algeria, Tunisia	Great Oriental Erg	4	32	100	200	Edmunds, W. M., Guendouz, A. H., Mamou, A., Moulla, A., Shand, P., Zouari, K. Groundwater evolution in the Continental Intercalaire aquifer of southern Algeria and Tunisia: trace element and isotopic indicators. Applied Geochemistry, 18, 805-822 (2003). Guendouz, A., Moulla,

Country	Aquifer	Lon.	Lat.	* Fossil groundwater depth (m)		Primary source of radiocarbon and tritium data
						A. S., Edmunds, W. M., Zouari, K., Shand, P., Mamou, A. Hydrogeochemical and isotopic evolution of water in the Complexe Terminal aquifer in the Algerian Sahara. Hydrogeology Journal, 11, 483-495 (2003). Abid, K., Dulinski, M., Ammar, F. H., Rozanski, K., Zouari, K. Deciphering interaction of regional aquifers in Southern Tunisia using hydrochemistry and isotopic tools. Applied Geochemistry, 27, 44-55 (2012). Abid, K., Zouari, K., Abidi, B. Identification and characterisation of hydrogeological relays of continental intercalaire aquifer of southern Tunisia. Carbonates Evaporites, 25, 65-75 (2010). Abid, K., Ammar, F. H., Chkir, N., Zouari, K. Relationship between Senonian and deep aquifers in Southern Tunisia. Quaternary International, 257, 13-26 (2012).
Estonia	Cambrian-Vendian	27	58	40	50	Vaikmäe, R., Vallner, L., Loosli, H. H., Blaser, P. C., Juillard-Tardent, M. Palaeogroundwater of glacial origin in the Cambrian-Vendian aquifer of northern Estonia. Geological Society, London, Special Publications, 189, 17-27 (2001).
France	Aquitaine Basin	0	46	80	180	Jiráková, H., Huneau, F., Celle-Jeanton, H., Hrkal, Z., Le Coustumer, P. Palaeorecharge conditions of the deep aquifers of the Northern Aquitaine region (France). Journal of Hydrology, 368, 1-16 (2009).
France	Bathonian-Bajocian	0	49	20	55	Barbecot, F., Marlin, C., Gibert, E., Dever, L. Hydrochemical and isotopic characterisation of the Bathonian and Bajocian coastal aquifer of the Caen area (northern France). Applied Geochemistry, 15, 791-805 (2000).
France	Lorraine Sandstone	7	48	120	320	Celle-Jeanton, H. Huneau, F., Travi, Y., Edmunds, W. M. Twenty years of groundwater evolution in the Triassic sandstone aquifer of Lorraine: Impacts on baseline water quality. Applied Geochemistry, 24, 1198-1213 (2009).
Hungary	Great Hungarian Plain	21	47	80	250	Stute, M., Deák, J. Environmental isotope study ^{14}C, ^{13}C, ^{18}O, D, noble gases on deep groundwater circulation systems in Hungary with reference to paleoclimate. Radiocarbon, 31, 902-918 (1989).
Hungary	Pannonian Basin	20	46	160	300	Varsányi, I., Palcsu, L., Kovács, L. Ó. Groundwater flow system as an archive of palaeotemperature: noble gas, radiocarbon, stable isotope and geochemical study in the Pannonian Basin, Hungary. Applied Geochemistry, 26, 91-104 (2011). Szocs, T., Rman, N., Süveges, M., Palcsu, L., Tóth, G., Lapanje, A. The application of isotope and chemical analyses in managing transboundary groundwater resources. Applied Geochemistry, 32, 95-107 (2013).
Indonesia	Jakarta Basin	107	−6	50	135	Geyh, M. A., Sofner, B. Groundwater analysis of environmental carbon and other isotopes from the Jakarta Basin Aquifer, Indonesia. Radiocarbon, 31, 919-925 (1989). Kagabu, M., Shimada, J., Delinom, R., Tsujimura, M., Taniguchi, M. Groundwater flow system under a rapidly urbanizing coastal city as determined by hydrogeochemistry. Journal of Asian Earth Sciences, 40, 226-239 (2011). Kagabu, M., Shimada, J., Delinom, R., Nakamura, T., Taniguchi, M. Groundwater age rejuvenation caused by excessive urban pumping in Jakarta area, Indonesia. Hydrogeological Processes, 27, 2591-2604 (2013).
Israel	Arava Valley	35	31	80	250	Vengosh, A., Hening, S., Ganor, J., Mayer, B., Weyhenmeyer, C. E., Bullen, T. D., Paytan, A. New isotopic evidence for the origin of groundwater from the Nubian Sandstone Aquifer in the Negev, Israel. Applied

Country	Aquifer	Lon.	Lat.	* Fossil groundwater depth (m)		Primary source of radiocarbon and tritium data
						Geochemistry, 22, 1052-1073 (2007). Burg, A., Zilberbrand, M., Yechieli, Y. Radiocarbon variability in groundwater in an extremely arid zone—the Arava Valley, Israel. Radiocarbon, 55, 963-978 (2013).
India	Tiruvadanai Aquifer	79	10	130	280	Kumar, U. S., Sharma, S., Navada, S. V., Deodhar, A. S. Environmental isotopes investigation on recharge processes and hydrodynamics of the coastal sedimentary aquifers of Tiruvadanai, Tamilnadu State, India. Journal of Hydrology, 364, 23-39 (2009).
Italy	Emilia Romagna Plain	11	45	110	300	Gorgni, C., Martinelli, G., Sighinolfi, G. P. Isotopic evidence of paleowaters in the Po sedimentary basin (Northern Italy). Geochemical Journal, 16, 51-61 (1982). Martinelli, G., Chahoud, A., Dadomo, A., Fava, A. Isotopic features of Emilia-Romagna region (North Italy) groundwaters: Environmental and climatological implications. Journal of Hydrology, 519, 1928-1938 (2014). Mayer, A., Sültenfuß, J., Travi, Y., Rebeix, R., Purtschert, R., Claude, C., Le Gal La Salle, C., Miche, H., Conchetto, E. A multi-tracer study of groundwater origin and transit-time in the aquifers of the Venice region (Italy). Applied Geochemistry, 50, 177-198 (2014).
Japan	Kimitsu-Yoro Aquifer	140	35	360	440	Tosaki, Y., Tase, N., Kondoh, A., Kimikazu, S., Takahasi, T., Nagashima, Y. Distribution of 36Cl in the Yoro River Basin, Central Japan, and its relation to the residence time of the regional groundwater flow system. Water, 3, 64-78 (2011). Machida, I., Suzuki, Y., Takeuchi, M. Carbon-14 age and chemical evolution of $Ca(HCO_3)_2$-type groundwater of age less than 8,000 years in a confined sandy and muddy Pleistocene aquifer, Japan. Hydrogeology Journal, 21, 1289-1305 (2013).
Libya	Kufra Basin	23	24	20	450	Al Faitouri, M., Sanford, W. E. Stable and radio-isotope analysis to determine recharge timing and paleoclimate of sandstone aquifers in central and southeast Libya. Hydrogeology Journal, DOI 10.1007/s10040-015-1232-7, (2015).
Libya	Sirte Basin	22	28	20	60	Edmunds, W. M., Wright, E. P. Groundwater recharge and palaeoclimate in the Sirte and Kufra basins, Libya. Journal of Hydrology, 40, 215-241 (1979).
Nigeria	Chad Basin	14	11	0	275	Maduabuchi, C., Faye, S., Maloszewski, P. Isotope evidence of palaeorecharge and palaeoclimate in the deep confined aquifers of the Chad Basin, NE Nigeria. Science of the Total Environment, 370, 467-479 (2006).
Poland	Malm Limestones	20	50	40	100	Zuber, A., Weise, S. M., Motyka, J., Osenbrück, K., Różański, K. Age and flow pattern of groundwater in a Jurassic limestone aquifer and related Tertiary sands derived from combined isotope, noble gas and chemical data. Journal of Hydrology, 286, 87-112 (2004).
Poland	Mazovian Basin	21	52	0	175	Zuber, A., Weise, S. M., Osenbrück, Pajnowska, H., Grabczak, J. Age and recharge pattern of water in the Oligocene of the Mazovian basin (Poland) as indicated by environmental tracers. Journal of Hydrology, 233, 174-188 (2000).
Poland	Southern Carbonates	21	50	160	190	Zuber, A., Weise, S. M., Osenbriick, K., Mateńko, T. Origin and age of saline waters in Busko Spa (Southern Poland) determined by isotope, noble gas and hydrochemical methods: evidence of interglacial and pre-Quaternary warm climate recharges. Applied Geochemistry, 12, 643-660 (1997). Samborska, K., Różkowski, A., Małoszewski, P.

Country	Aquifer	Lon.	Lat.	* Fossil groundwater depth (m)		Primary source of radiocarbon and tritium data
						Estimation of groundwater residence time using environmental radioisotopes (^{14}C, T) in carbonate aquifers, southern Poland. Isotopes in environmental and health studies, 49, 73-97 (2012).
Senegal	Diass Aquifer	−15	16	50	300	Castany, G., Marce, A., Margat, J., Moussu, H., Vuillaume, Y., Evin, J. An environmental isotope study of the groundwater regime in large aquifers. in: Isotope techniques in groundwater hydrology 1974, Vol. I., International Atomic Energy Agency, Vienna, Austria, 243-256 (1974). Madioune, D. H., Faye, S., Orban, P., Brouyère, S., Dassargues, A., Mudry, J., Stumpp, C., Maloszewski, P. Application of isotopic tracers as a tool for understanding hydrodynamic behavior of the highly exploited Diass aquifer system (Senegal). Journal of Hydrology, 511, 443-459 (2014).
Thailand	Bangkok Basin	101	14	60	120	Sanford, W., Buapeng, S. Assessment of a groundwater flow model of the Bangkok Basin, Thailand, using carbon-14-based ages and paleohydrology. Hydrogeology Journal, 4, 26-40 (1996).
Tunisia	Djérid Basin	8	34	30	120	Kamel, S. Dassi, L., Zouari, K. Approche hydrogéologique et hydrochimique des échanges hydrodynamiques entre aquifères profond et superficiel du bassin du Djérid, Tunisie. Hydrological Sciences Journal, 51, 713-730 (2006).
Turkey	Kazan Trona	40	33	20	70	Arslan, S., Yazicigil, H., Stute, M., Schlosser, P. Environmental isotopes and noble gases in the deep aquifer system of Kazan Trona Ore Field, central Turkey and links to paleoclimate. Quaternary Research, 79, 292-303 (2012).
USA	California Bay: East	−122	38	125	200	Water Quality Portal (www.waterqualitydata.us)
USA	California Bay: Monterey	−122	37	190	250	Water Quality Portal (www.waterqualitydata.us)
USA	Cambrian-Ordovician	−88	42	260	400	Water Quality Portal (www.waterqualitydata.us)
USA	Castle-Hayne Aquifer	−78	35	55	80	Kennedy, C.D., Genereux, D.P. ^{14}C groundwater age and the importance of chemical fluxes across aquifer boundaries in confined Cretaceous aquifers of North Carolina, USA. Radiocarbon, 49, 1181-1203 (2007). Water Quality Portal (www.waterqualitydata.us)
USA	Columbia Basin	−119	47	50	350	Brown, K. B., McIntosh, J. C., Rademacher, L. K., Lohse, K. A. Impacts of agricultural irrigation recharge on groundwater quality in a basalt aquifer system (Washington, USA): a multi-tracer approach. Hydrogeology Journal, 19, 1039-1051 (2011). Water Quality Portal (www.waterqualitydata.us)
USA	Denver Basin	−105	39	125	200	Water Quality Portal (www.waterqualitydata.us)
USA	Eureka Coast	−124	41	70	120	Water Quality Portal (www.waterqualitydata.us)
USA	Floridan Aquifer	−82	33	90	140	Clark, J. F., Stute, M., Schlosser, P., Drenkard, S., Bonani, G. A tracer study of the Floridan aquifer in southeastern Georgia: Implications for groundwater flow and paleoclimate. Water Resources Research, 33, 281-289 (1997). Water Quality Portal (www.waterqualitydata.us)
USA	Central High Plains	−103	37	120	190	Clark, J. F., Davisson, M. L., Hudson, G. B., Macfarlane, P. A. Noble gases, stable isotopes, and radiocarbon as tracers of flow in the Dakota aquifer, Colorado and Kansas. Journal of Hydrology, 211, 151-167 (1998). Dutton, A. R. Groundwater isotopic evidence for paleorecharge in US High Plains aquifers. Quaternary Research, 43, 221-231 (1995).

Country	**Aquifer**	**Lon.**	**Lat.**	*** Fossil groundwater depth (m)**		**Primary source of radiocarbon and tritium data**
						McMahon, P. B., Böhlke, J. K., Christenson, S. C. Geochemistry, radiocarbon ages, and paleorecharge conditions along a transect in the central High Plains aquifer, southwestern Kansas, USA. Applied Geochemistry, 19, 1655-1686 (2004). Water Quality Portal (www.waterqualitydata.us)
USA	North High Plains	−104	42	170	240	McMahon, P. B., Böhlke, J. K., Carney, C. P. Vertical Gradients in Water Chemistry and Age in the Northern High Plains Aquifer, Nebraska, 2003. US Geological Survey Scientific Investigations Report 2006–5294 (2006). Water Quality Portal (www.waterqualitydata.us)
USA	South High Plains	−102	36	120	275	Water Quality Portal (www.waterqualitydata.us)
USA	Houston Coast (TX)	−95	29	20	140	Water Quality Portal (www.waterqualitydata.us)
USA	Jacksonville Aquifer	−82	30	55	70	Water Quality Portal (www.waterqualitydata.us)
USA	Livermore Valley	−122	38	200	220	Water Quality Portal (www.waterqualitydata.us)
USA	Los Angeles Basin	−118	34	360	550	Swarzenski, P. W., Baskaran, M., Rosenbauer, R. J., Edwards, B. D., Land, M. A combined radio-and stable-isotopic study of a California coastal aquifer system. Water, 5, 480-504 (2013). Water Quality Portal (www.waterqualitydata.us)
USA	Tertiary Aquifer (MT)	−105	47	20	55	Water Quality Portal (www.waterqualitydata.us)
USA	Mahomet Aquifer	−89	40	100	460	Hackley, K. C., Panno, S. V., Anderson, T. F. Chemical and isotopic indicators of groundwater evolution in the basal sands of a buried bedrock valley in the midwestern United States: Implications for recharge, rock-water interactions, and mixing. Geological Society of America Bulletin, 122, 1047-1066 (2010). Water Quality Portal (www.waterqualitydata.us)
USA	Aquia-Patapsco Aquifer	−77	39	25	75	Aeschbach-Hertig, W., Stute, M., Clark, J. F., Reuter, R. F., Schlosser, P. A paleotemperature record derived from dissolved noble gases in groundwater of the Aquia Aquifer (Maryland, USA). Geochimica et Cosmochimica Acta, 66, 797-817 (2002). Plummer, L. N., Eggleston, J. R., Andreasen, D. C., Raffensperger, J. P., Hunt, A. G., Castle, G. C. Old groundwater in parts of the upper Patapsco aquifer, Atlantic Coastal Plain, Maryland, USA: evidence from radiocarbon, chlorine-36 and helium-4. Hydrogeology Journal, 20, 1269-1294 (2012). Water Quality Portal (www.waterqualitydata.us)
USA	Mississippi Alluvial	−91	33	105	135	Water Quality Portal (www.waterqualitydata.us)
USA	Mississippi Embayment	−93	33	45	115	Water Quality Portal (www.waterqualitydata.us)
USA	Mississippi Embayment: Memphis	−90	35	195	510	Larsen, D., Gentry, R. W., Solomon, D. K. The geochemistry and mixing of leakage in a semi-confined aquifer at a municipal well field, Memphis, Tennessee, USA. Applied geochemistry, 18, 1043-1063 (2003). Water Quality Portal (www.waterqualitydata.us)
USA	Atlantic Coast	−77	36	40	80	Water Quality Portal (www.waterqualitydata.us)
USA	Palouse Basin	−117	47	60	80	Douglas, A., Osiensky, J. L., Keller, C. K. Carbon-14 dating of ground water in the Palouse Basin of the Columbia river

Country	Aquifer	Lon.	Lat.	* Fossil groundwater depth (m)		Primary source of radiocarbon and tritium data
						basalts. Journal of Hydrology, 334, 502-512 (2007). Water Quality Portal (www.waterqualitydata.us)
USA	San Diego Basin	−117	33	180	250	Water Quality Portal (www.waterqualitydata.us)
USA	Silurian-Devonian (IL)	−88	41	30	430	Water Quality Portal (www.waterqualitydata.us)
USA	South Central Valley (CA)	−121	38	240	280	Jurgens, B. C., Burow, K. R., Dalgish, B. A., Shelton, J. L. Hydrogeology, water chemistry, and factors affecting the transport of contaminants in the zone of contribution of a public-supply well in Modesto, eastern San Joaquin Valley, California. Scientific Investigations Report 2008–5156 (2008). Water Quality Portal (www.waterqualitydata.us)
USA	Surficial Aquifers	−81	27	60	110	Morrissey, S. K., Clark, J. F., Bennett, M., Richardson, E., Stute, M. Groundwater reorganization in the Floridan aquifer following Holocene sea-level rise. Nature Geoscience, 3, 683-687 (2010). Water Quality Portal (www.waterqualitydata.us)
USA	Tampa Bay	−82	27	25	50	Water Quality Portal (www.waterqualitydata.us)
USA	Temecula-Murrieta Basin	−117	33	305	330	Water Quality Portal (www.waterqualitydata.us)
USA	Wharton Tract	−75	40	70	140	Thompson, G. M., Hayes, J. M. Trichlorofluoromethane in groundwater—a possible tracer and indicator of groundwater age. Water Resources Research, 15, 546-554 (1979). Water Quality Portal (www.waterqualitydata.us)

* the depth to fossil groundwater below the land surface. A shallow and deep estimate are provided. The shallower depth to fossil groundwater (left column under heading “Fossil groundwater (m)”) specifies the depth below which the majority (>50%) of well water samples from a given aquifer must contain some fraction of fossil groundwater (i.e., over half the samples have a minimum fossil groundwater fraction of greater than zero). The deeper depth to fossil groundwater (right column under heading “Fossil groundwater (m)”) represents a depth below which the majority (>50%) of sampled well waters from a given aquifer system contain mostly fossil groundwater (i.e., over half of the samples deeper than the depth have a minimum fossil groundwater fraction of greater than 0.5).

Table S2. Groundwater isotope data sources outside of the study aquifers

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Table S3. Industrial, agricultural, domestic or municipal well construction purposes

State	Well construction purposes included in our analysis shown in Figure 3
California	[Other Recreation] [Water Supply Irrigation - Landscape] [Water Supply Domestic] [Water Supply Public] [Other Fire or Frost Protection] [Other Bottled Water] [Other Power Generation] [Water Supply Industrial] [Other Dairy Supply] [Other iriigation] [Other Irrigation] [Water Supply Irrigation - Agriculture] [Water Supply Stock or Animal Watering] [Water Supply Irrigation Agricultural]
Colorado	[/DOMESTIC] [ALL BENEFICIAL USES/DOMESTIC] [ALL BENEFICIAL USES/MUNICIPAL] [AUGMENTATION/DOMESTIC] [DOMESTIC/] [DOMESTIC/ALL BENEFICIAL USES] [DOMESTIC/FIRE] [DOMESTIC/MUNICIPAL] [DOMESTIC/OTHER] [DOMESTIC/RECREATION] [FIRE/] [FIRE/DOMESTIC] [HOUSEHOLD USE ONLY/] [HOUSEHOLD USE ONLY/DOMESTIC] [HOUSEHOLD USE ONLY/FIRE] [HOUSEHOLD USE ONLY/OTHER] [HOUSEHOLD USE ONLY/RECREATION] [MUNICIPAL/] [MUNICIPAL/ALL BENEFICIAL USES] [MUNICIPAL/DOMESTIC] [MUNICIPAL/FIRE] [MUNICIPAL/OTHER] [MUNICIPAL/RECREATION] [OTHER/DOMESTIC] [OTHER/FIRE] [RECREATION/] [RECREATION/DOMESTIC] [RECREATION/OTHER] [HOUSEHOLD USE ONLY/AUGMENTATION] [MUNICIPAL/AUGMENTATION] [DOMESTIC/AUGMENTATION] [/COMMERCIAL] [/INDUSTRIAL] [ALL BENEFICIAL USES/COMMERCIAL] [COMMERCIAL/] [COMMERCIAL/AUGMENTATION] [COMMERCIAL/INDUSTRIAL] [COMMERCIAL/OTHER] [INDUSTRIAL/] [INDUSTRIAL/ALL BENEFICIAL USES] [INDUSTRIAL/COMMERCIAL] [INDUSTRIAL/OTHER] [INDUSTRIAL/POWER GENERATION] [OTHER/INDUSTRIAL] [COMMERCIAL/SNOW MAKING] [SNOW MAKING/] [SNOW MAKING/OTHER] [INDUSTRIAL/AUGMENTATION] [AUGMENTATION/IRRIGATION] [DOMESTIC/FISHERY] [FISHERY/IRRIGATION] [FISHERY/STOCK] [IRRIGATION/] [IRRIGATION/ALL BENEFICIAL USES] [IRRIGATION/FISHERY] [IRRIGATION/OTHER] [IRRIGATION/STOCK] [OTHER/STOCK] [RECREATION/STOCK] [STOCK/] [STOCK/FISHERY] [STOCK/IRRIGATION] [STOCK/OTHER] [IRRIGATION/AUGMENTATION] [FISHERY/] [COMMERCIAL/DOMESTIC] [COMMERCIAL/FIRE] [COMMERCIAL/HOUSEHOLD USE ONLY] [COMMERCIAL/MUNICIPAL] [COMMERCIAL/RECREATION] [DOMESTIC/COMMERCIAL] [DOMESTIC/INDUSTRIAL] [DOMESTIC/POWER GENERATION] [DOMESTIC/SNOW MAKING] [HOUSEHOLD USE ONLY/COMMERCIAL] [HOUSEHOLD USE ONLY/INDUSTRIAL] [INDUSTRIAL/DOMESTIC] [INDUSTRIAL/MUNICIPAL] [INDUSTRIAL/RECREATION] [MUNICIPAL/COMMERCIAL] [MUNICIPAL/INDUSTRIAL] [MUNICIPAL/SNOW MAKING] [RECREATION/POWER GENERATION] [DOMESTIC/IRRIGATION] [DOMESTIC/STOCK] [FIRE/IRRIGATION] [FIRE/STOCK] [FISHERY/] [FISHERY/RECREATION] [HOUSEHOLD USE ONLY/IRRIGATION] [HOUSEHOLD USE ONLY/STOCK] [INDUSTRIAL/STOCK] [IRRIGATION/DOMESTIC] [IRRIGATION/FIRE] [IRRIGATION/MUNICIPAL] [IRRIGATION/RECREATION] [MUNICIPAL/IRRIGATION] [MUNICIPAL/STOCK] [RECREATION/FISHERY] [RECREATION/IRRIGATION] [STOCK/DOMESTIC] [STOCK/FIRE] [STOCK/HOUSEHOLD USE ONLY] [STOCK/MUNICIPAL] [DOMESTIC/IRRIGATION] [FIRE/IRRIGATION] [HOUSEHOLD USE ONLY/IRRIGATION] [HOUSEHOLD USE] [ONLY/STOCK] [FIRE/STOCK] [FISHERY/RECREATION] [COMMERCIAL/FISHERY] [COMMERCIAL/IRRIGATION] [COMMERCIAL/STOCK] [INDUSTRIAL/IRRIGATION] [IRRIGATION/COMMERCIAL] [IRRIGATION/INDUSTRIAL] [IRRIGATION/SNOW MAKING] [STOCK/COMMERCIAL] [STOCK/INDUSTRIAL] [INDUSTRIAL/STOCK]
Kansas	[Pond/Swimming Pool/Recreation] [Public Water Supply] [Lawn and Garden - domestic only] [Domestic] [Domestic, changed from Irrigation] [Domestic, changed from Oil Field Water Supply] [Industrial] [Oil Field Water Supply] [Irrigation] [Feedlot] [Feedlot/Livestock/Windmill] [Domestic, Livestock]
Nebraska	[Domestic] [Commercial/Industrial] [Aquaculture] [Pit - Irrigation] [Irrigation] [Livestock]
Wyoming	[DOM_GW] [MUN_GW] [DOM_GW; MIS] [DOM_GW;MIS] [DOM_GW; TST] [MIS; MUN_GW] [DOM_GW; MUN_GW] [MIS; DOM_GW] [DOM_GW; MON] [IND_GW] [IND_GW; MIS] [IND_GW; MON] [IND_GW;MIS] [IND_GW; MIS; MON] [IND_GW; MIS; MIS] [MIS; IND_GW] [STK] [MIS; STK] [IRR_GW] [IRR_GW; STK] [STK; IRR_GW] [MON; STK] [IRR_GW; MIS] [MIS; MON; STK] [IRR_GW;MIS] [IRR_GW; MIS; STK] [MIS; IRR_GW] [STK; MIS] [IRR_GW;STK] [STK; MIS; MIS] [MIS; STK; IRR_GW] [MIS;STK] [STK; IRR_GW; MIS] [IRR_GW; MIS; MIS; MIS; MIS] [IRR_GW;MIS;STK] [DOM_GW; IND_GW] [DOM_GW; IND_GW; MIS] [DOM_GW; IND_GW; MUN_GW] [IND_GW; MUN_GW] [IND_GW; MIS; MUN_GW]

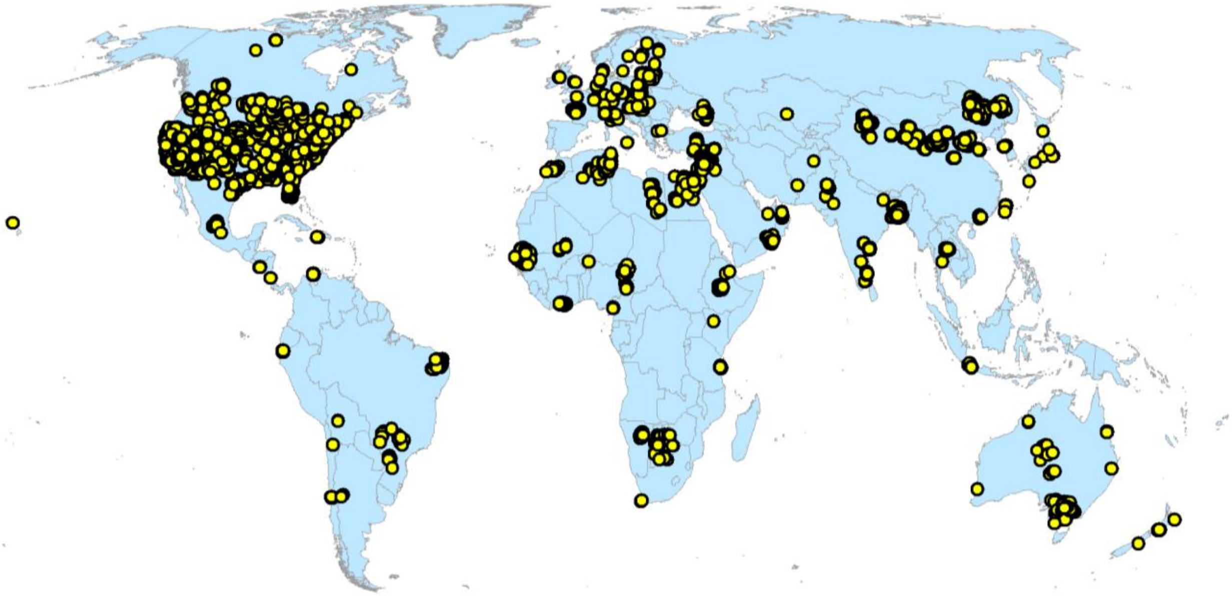


Figure S1. Locations of the analysed well waters with tritium and/or radiocarbon data (yellow circles). Outlines of countries are marked by grey lines.

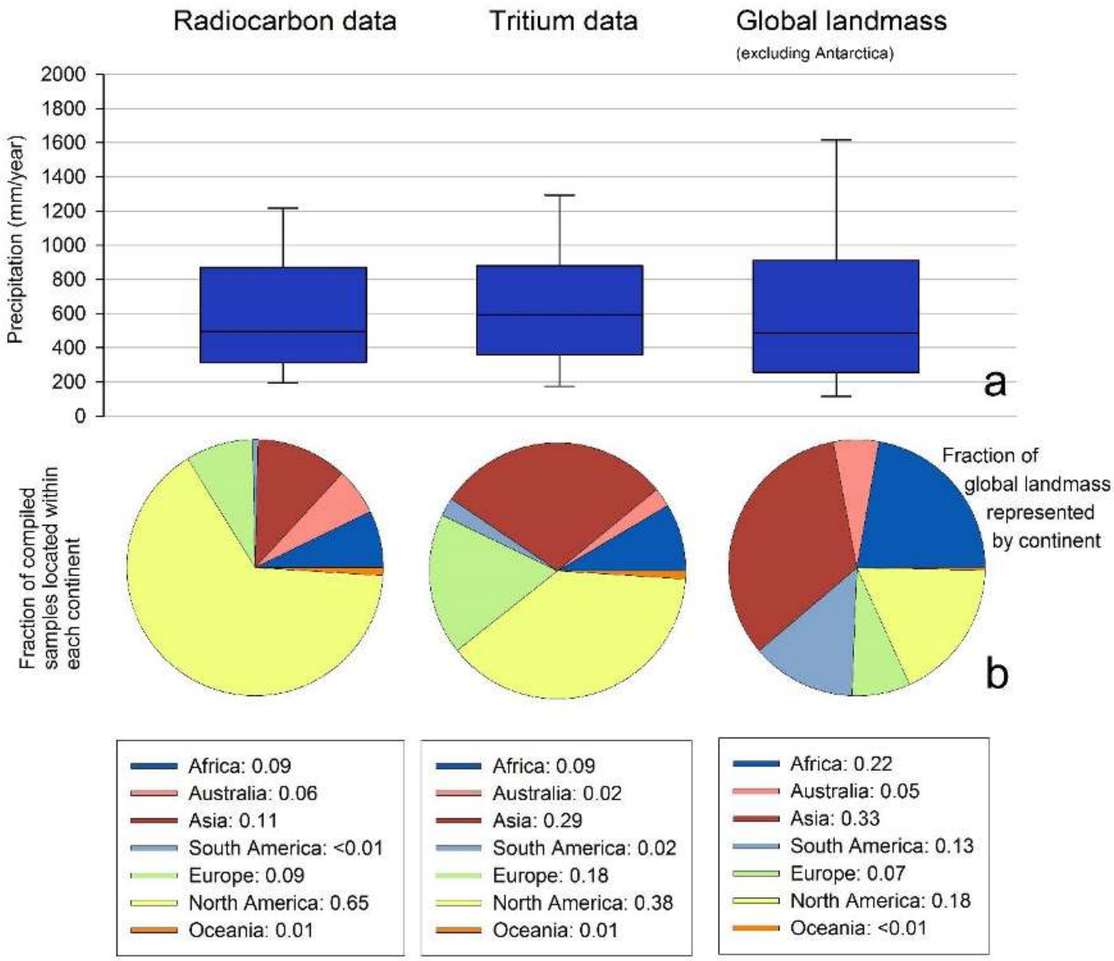


Figure S2. Sampling biases of radiocarbon (left column) and tritium (middle column), relative to the distribution of the global landmass by continent (right column). Panel (a) shows a box-whisker plot of annual precipitation (mm/year) at the wells where we compiled radiocarbon data (leftmost plot), the wells where we compiled tritium data (middle plot), and the entire global landmass excluding Antarctica (rightmost plot; data from Ref. 45). Panel (b) shows the fraction of all radiocarbon (leftmost pie chart) and tritium (middle pie chart) measurements that we compiled, juxtaposed against the distribution of landmass categorized by continental area (rightmost plot). Our dataset disproportionately represents some continents relative to their land areas (e.g., North America, yellow above). Our compiled data represent many climate zones and generally represents the global distribution of precipitation rates (panel a). However, we note that several continents are disproportionately represented in our compilation relative to the areas of the continents, specifically for North America, which accounts for 65% and 38% of compiled radiocarbon and tritium data, but only 18% of the global landmass.

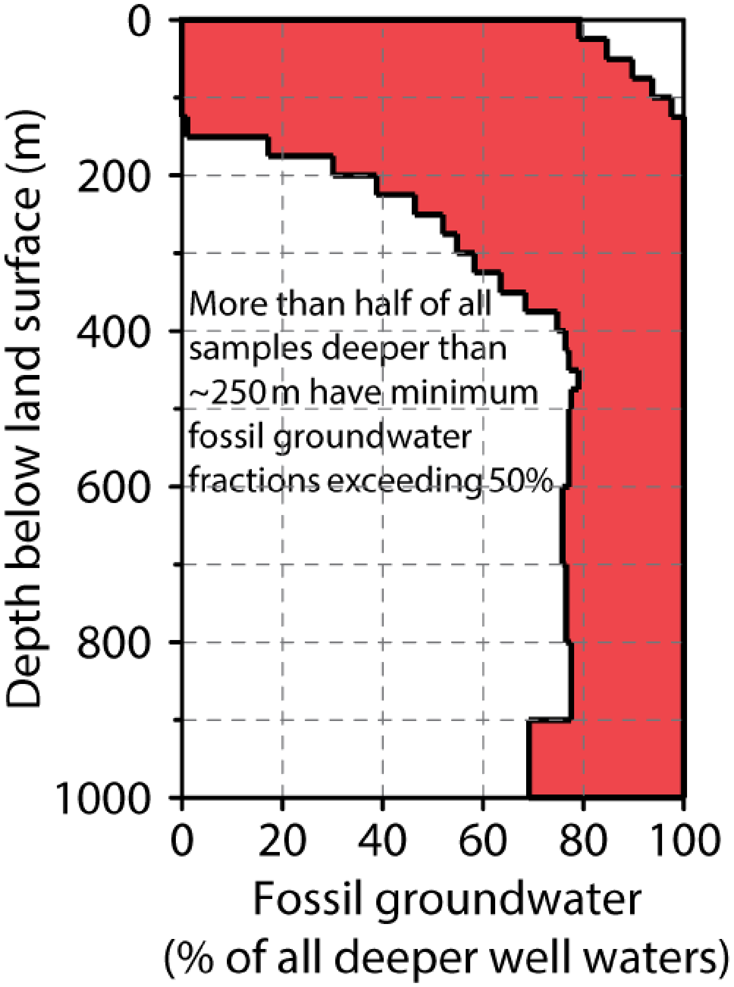
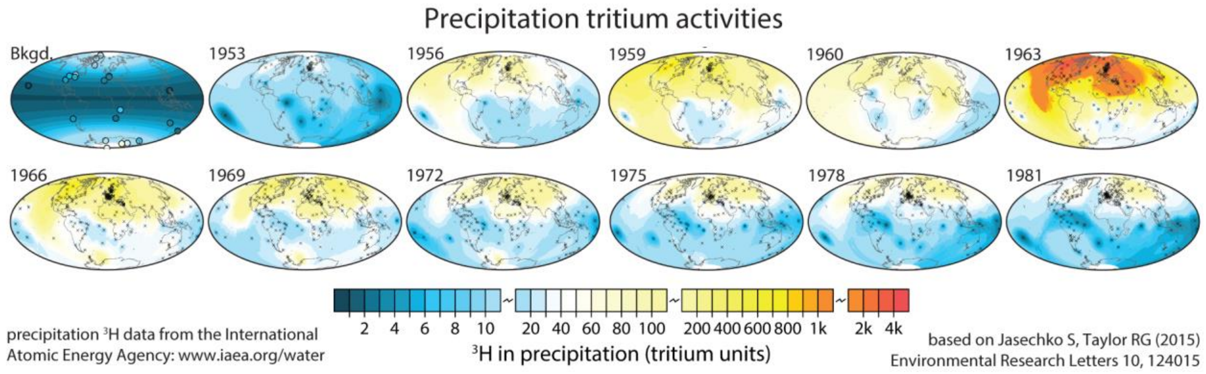


Figure S3. Depth variations of the fraction of fossil groundwater in all compiled underlying (i.e., deeper) wells. The figure presents the median value of the upper and lower (maximum and minimum) estimates of fossil and post-1953 groundwater as calculated using all underlying wells. More than half of all compiled well waters pumped from depths of deeper than 250 m below the land surface have a minimum fossil groundwater fraction of greater than 50%.


Precipitation tritium activities

precipitation ^{3}H data from the International Atomic Energy Agency: www.iaea.org/water

based on Jasechko S, Taylor RG (2015) Environmental Research Letters 10, 124015

Figure S4. Time series of interpolated global precipitation tritium from pre-1953 (background: “Bkgd.”) through the year 1981 (lower right plot). Precipitation tritium measurements are from the International Atomic Energy Agency’s precipitation isotope database (www.iaea.org/water). The maps shown here are based on those from Ref. 24.

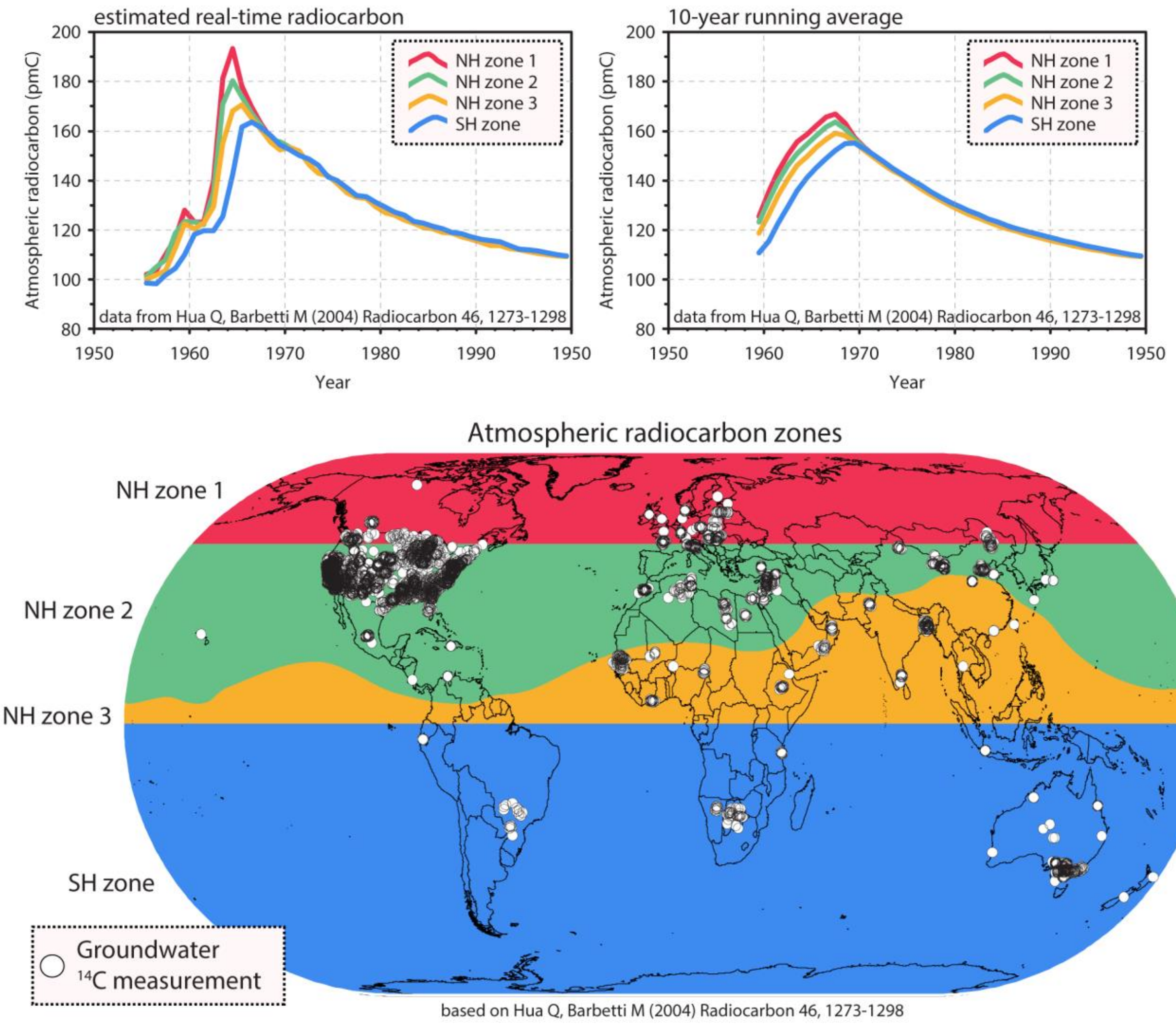


Figure S5. Atmospheric radiocarbon concentrations and groundwater 14C sampling locations. The upper two panels show estimated time series of atmospheric radiocarbon in three geographic zones (left panel), and a 10-year moving average of these atmospheric 14C data (right panel). That is, the data shown in the upper left panel are estimates of the real-time atmospheric radiocarbon content, and the line plots in the top right panel are 10-year moving averages of the data in the upper left panel. The lower map shows the locations of each of the radiocarbon zones, where the colours correspond to the colours of the lines in the upper two panels. The circles show groundwater well locations where fossil groundwater calculations were performed in our manuscript. The atmospheric radiocarbon time series and zones are based on Hua, Q. & Barbetti, M. (2004). Radiocarbon 46, 1273-1298 (Ref. 43).

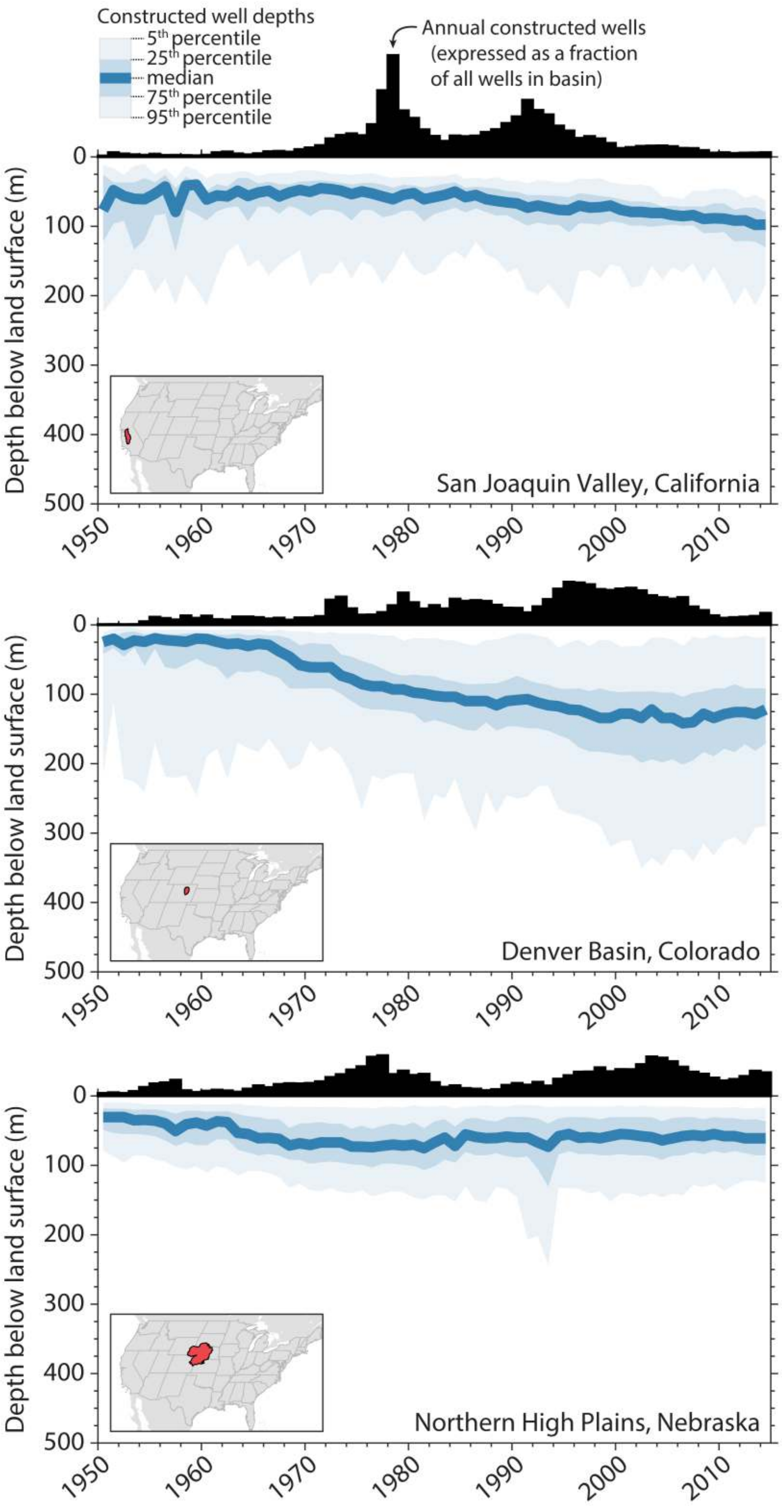
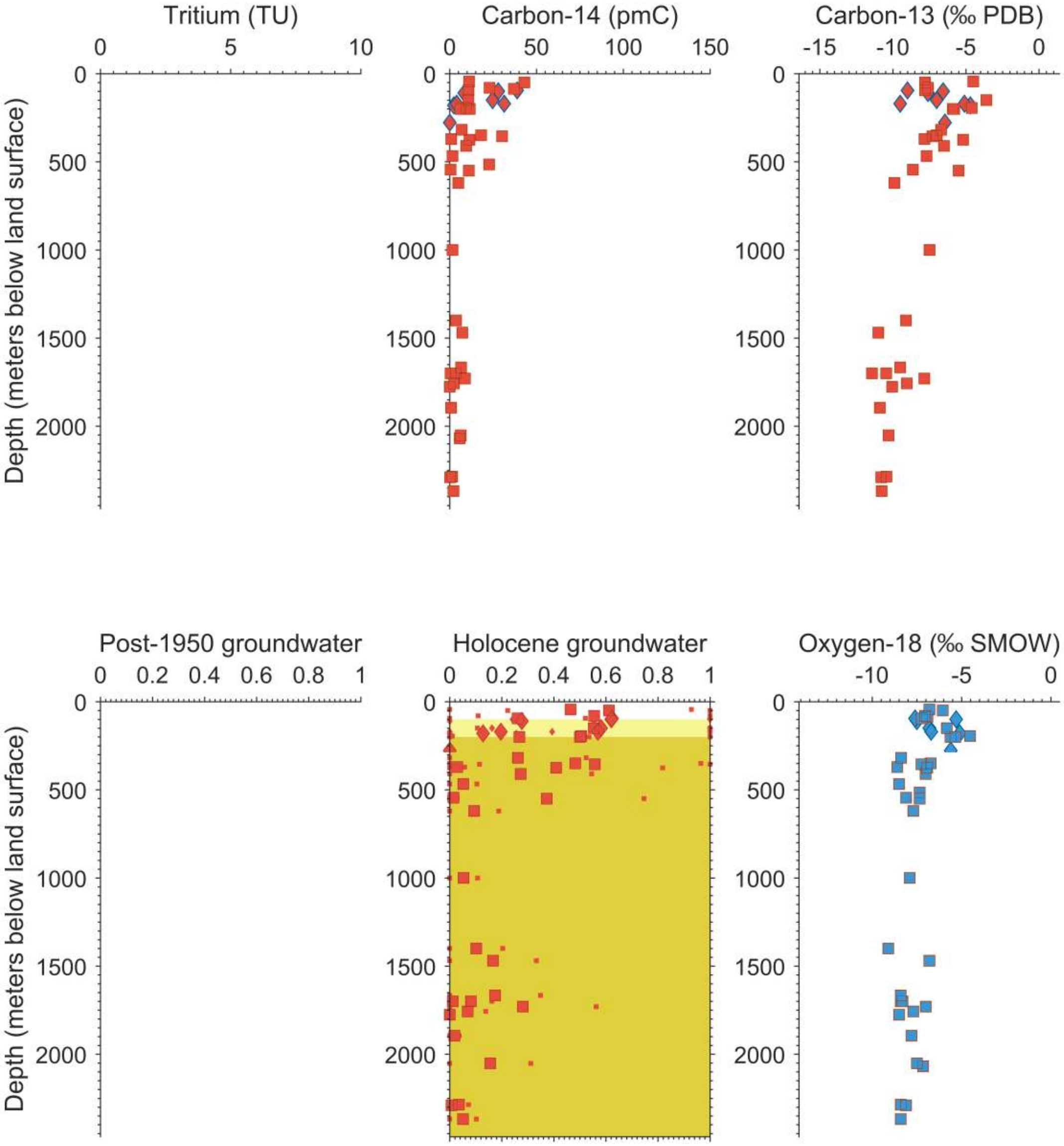
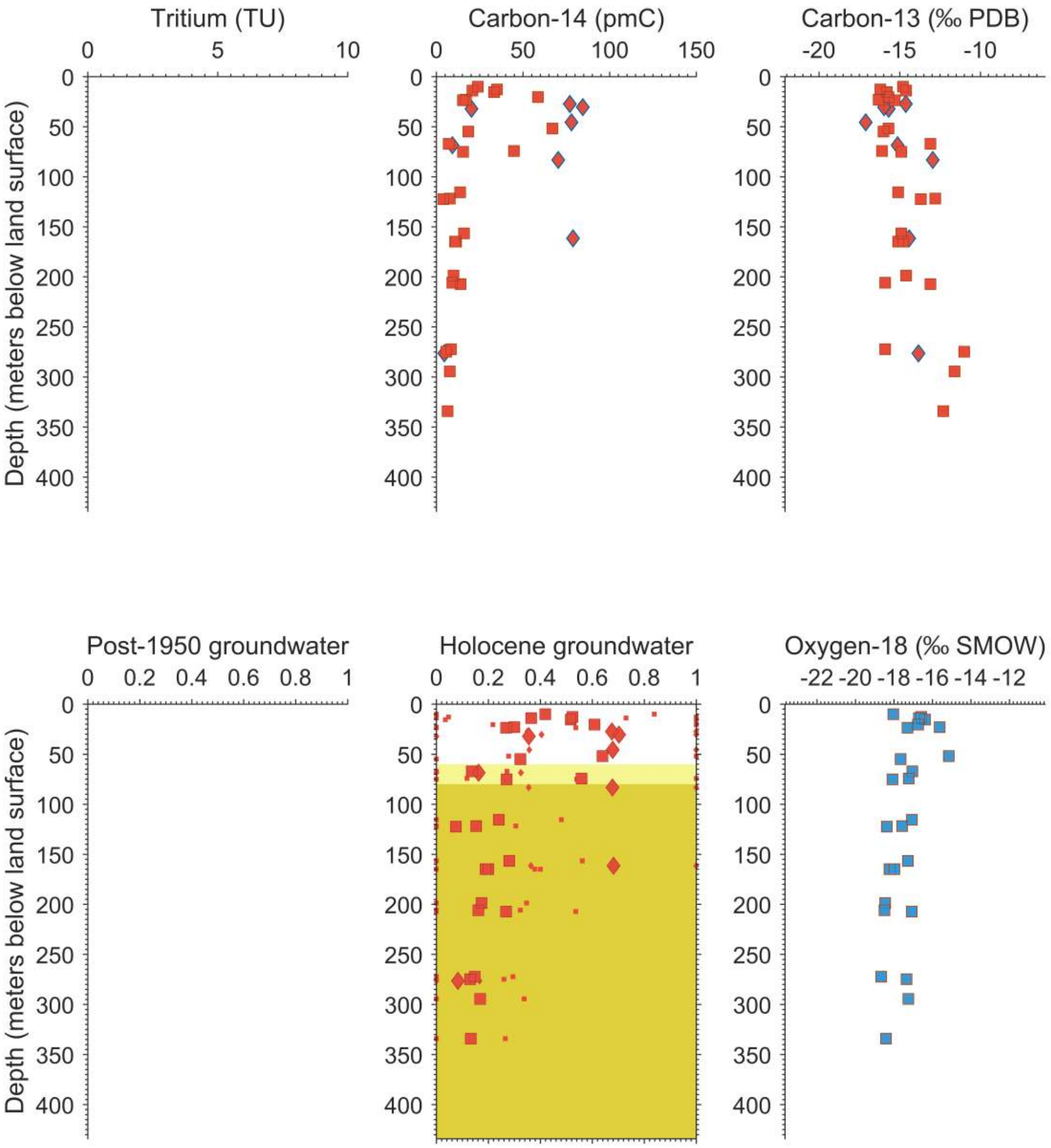


Figure S6. Groundwater well construction depth trends (1950-2015) in six western US aquifers. The blue line shows the annual median well construction depth, the medium blue shading marks the upper-lower quartile well depths, and the light blue shading marks the 5th-95th percentile range of wells. In the San Joaquin Valley and the Northern High Plains, constructed wells lie above the depth that these aquifer systems transition to mostly-fossil groundwater. In the Denver Basin, many wells are constructed at similar depths as fossil groundwaters.

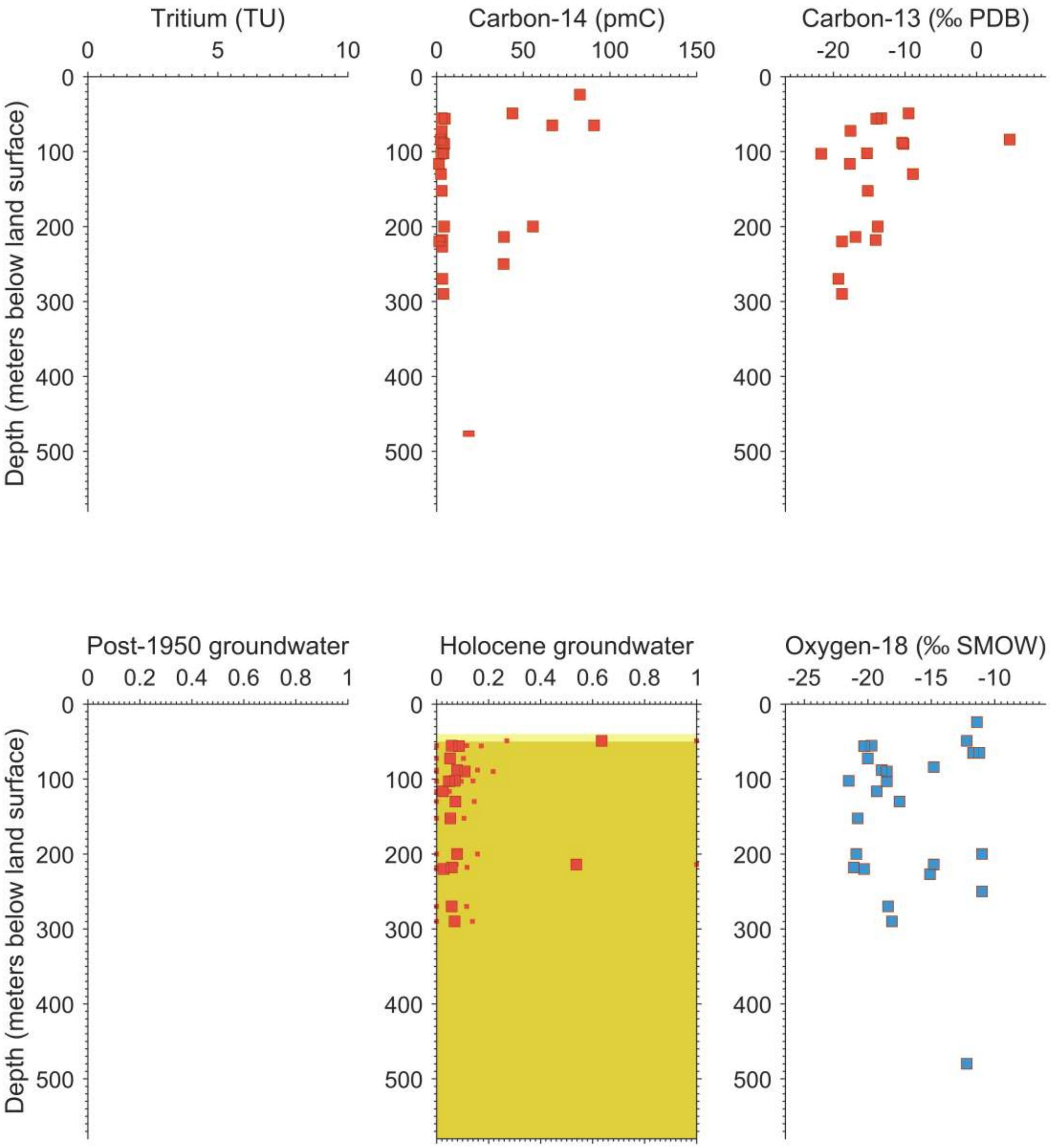
Figures S7 (following pages). For figure description see Supplementary Information section S2.



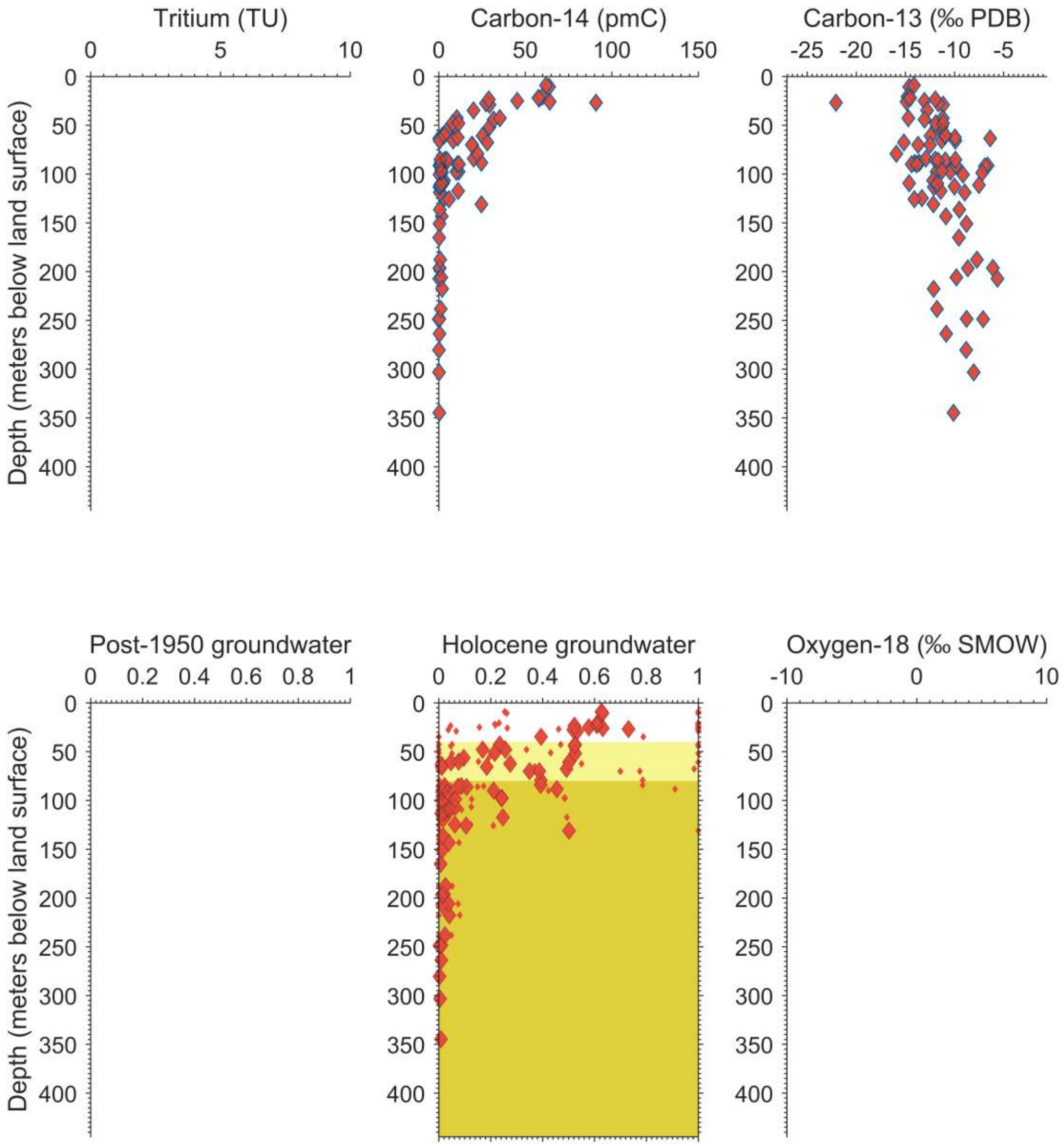
Great Oriental Erg: Continental Intercalaire, Algeria



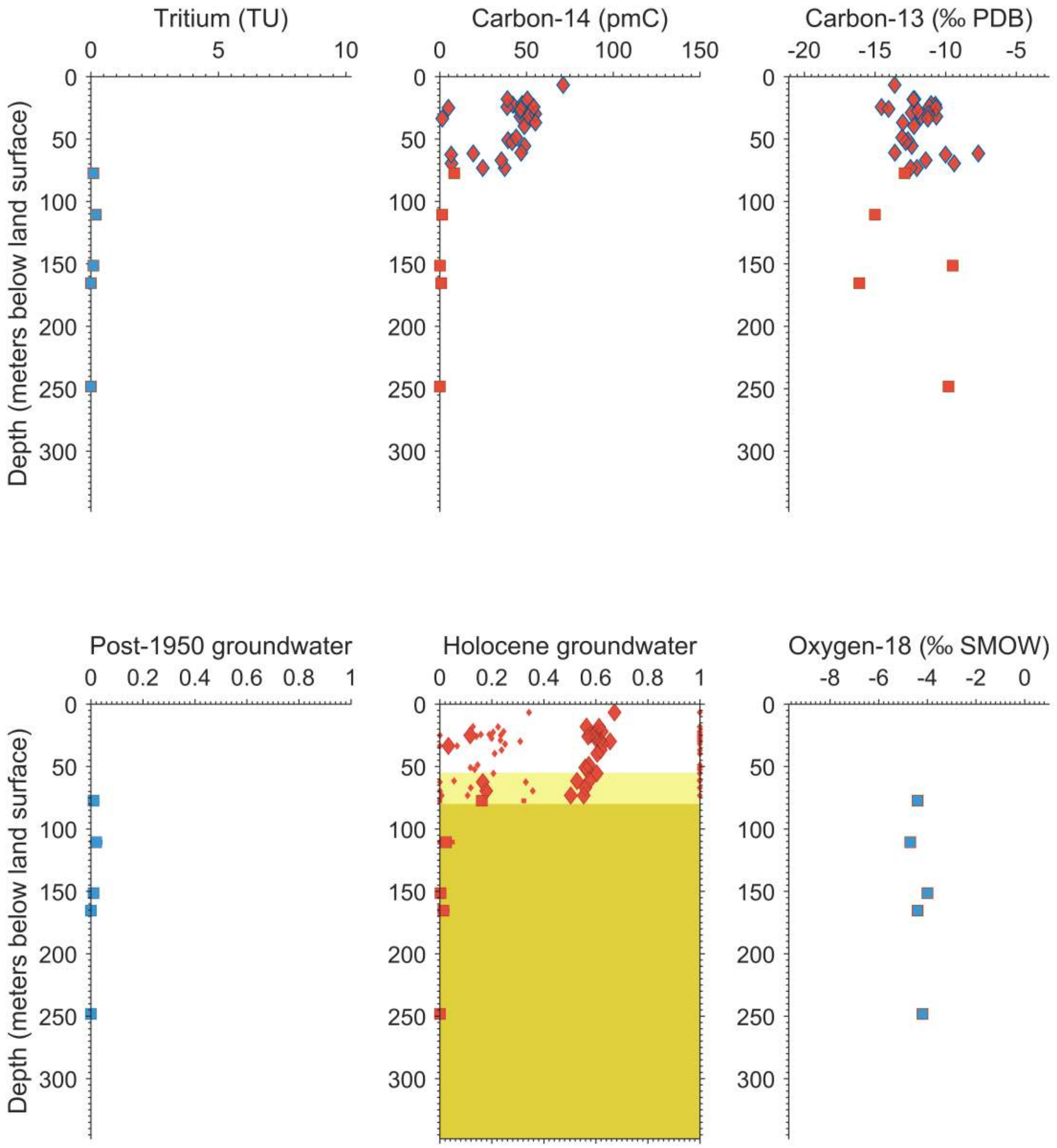
Palouse Basin, USA



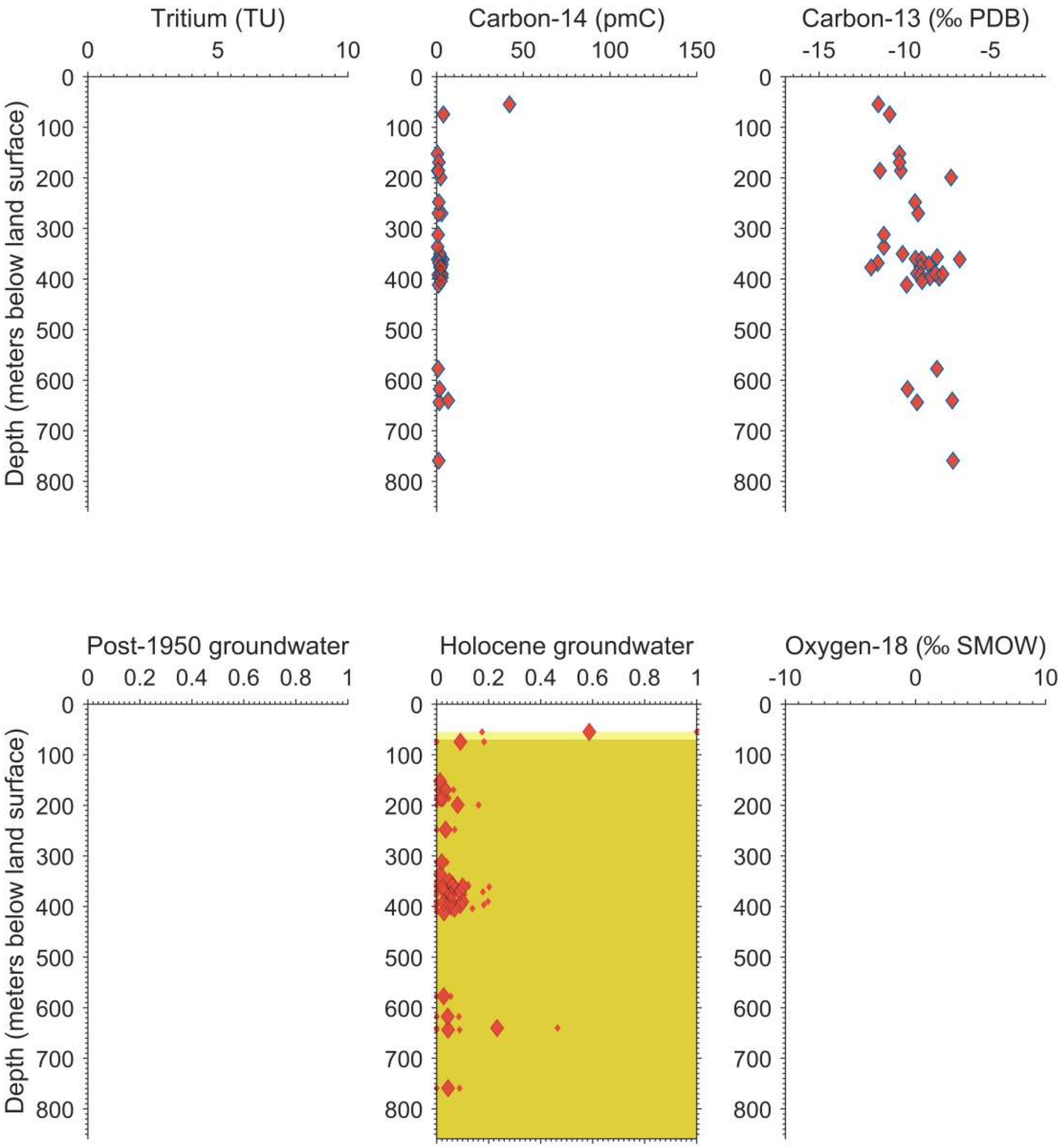
Cambrian-Vendian Aquifer, Estonia



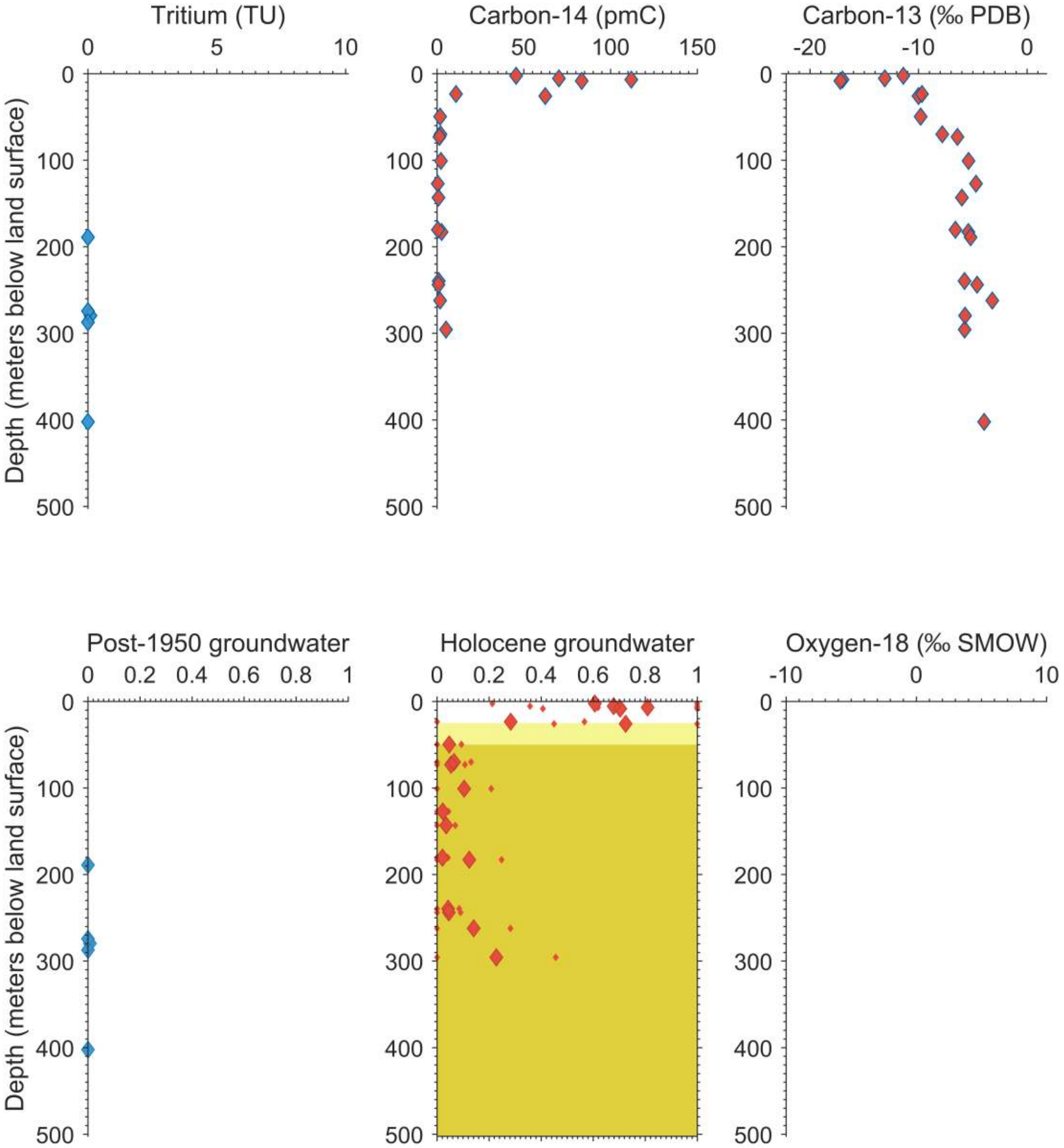
Northern Atlantic Coast, USA



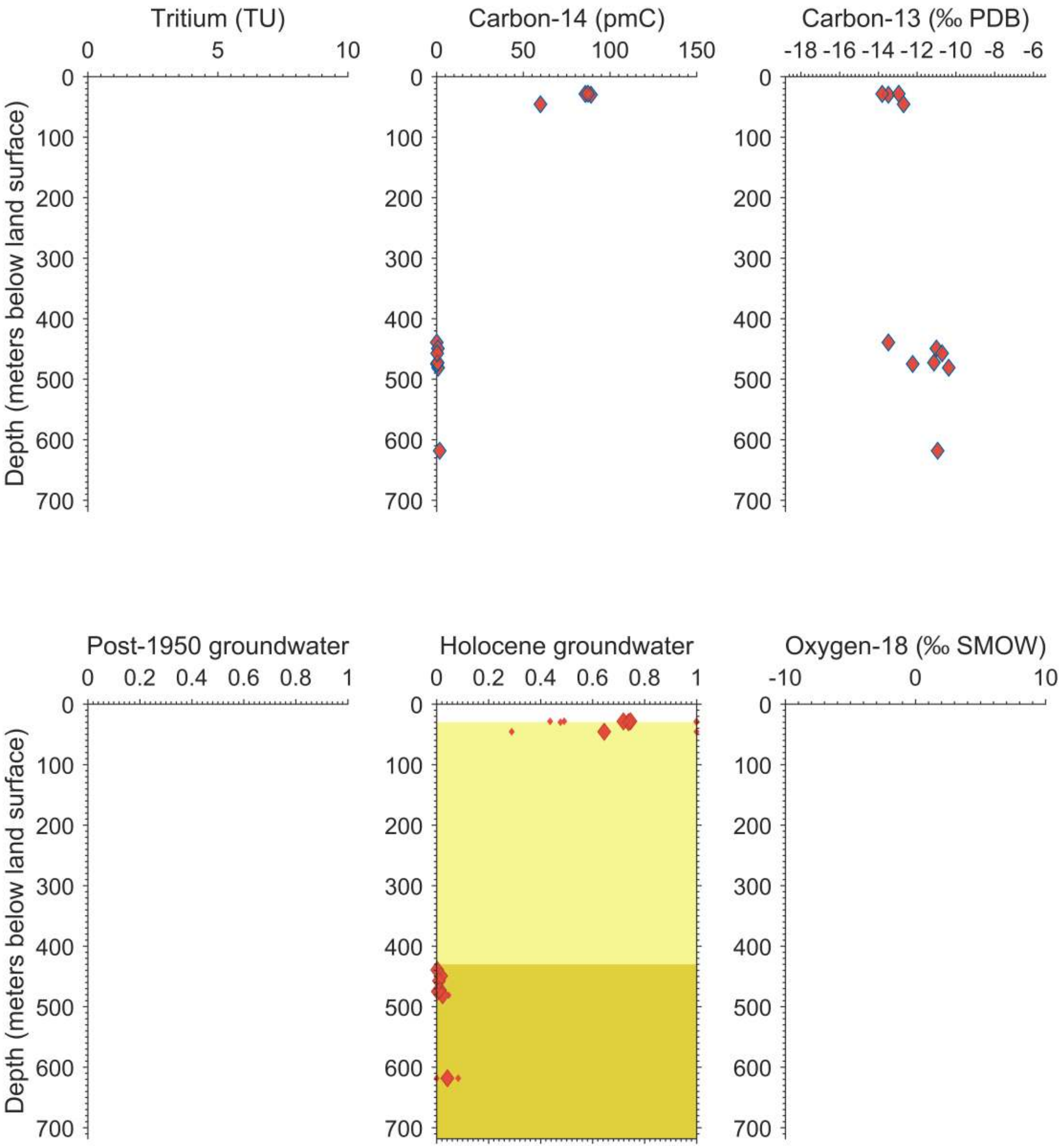
Castle Hayne Aquifer, USA



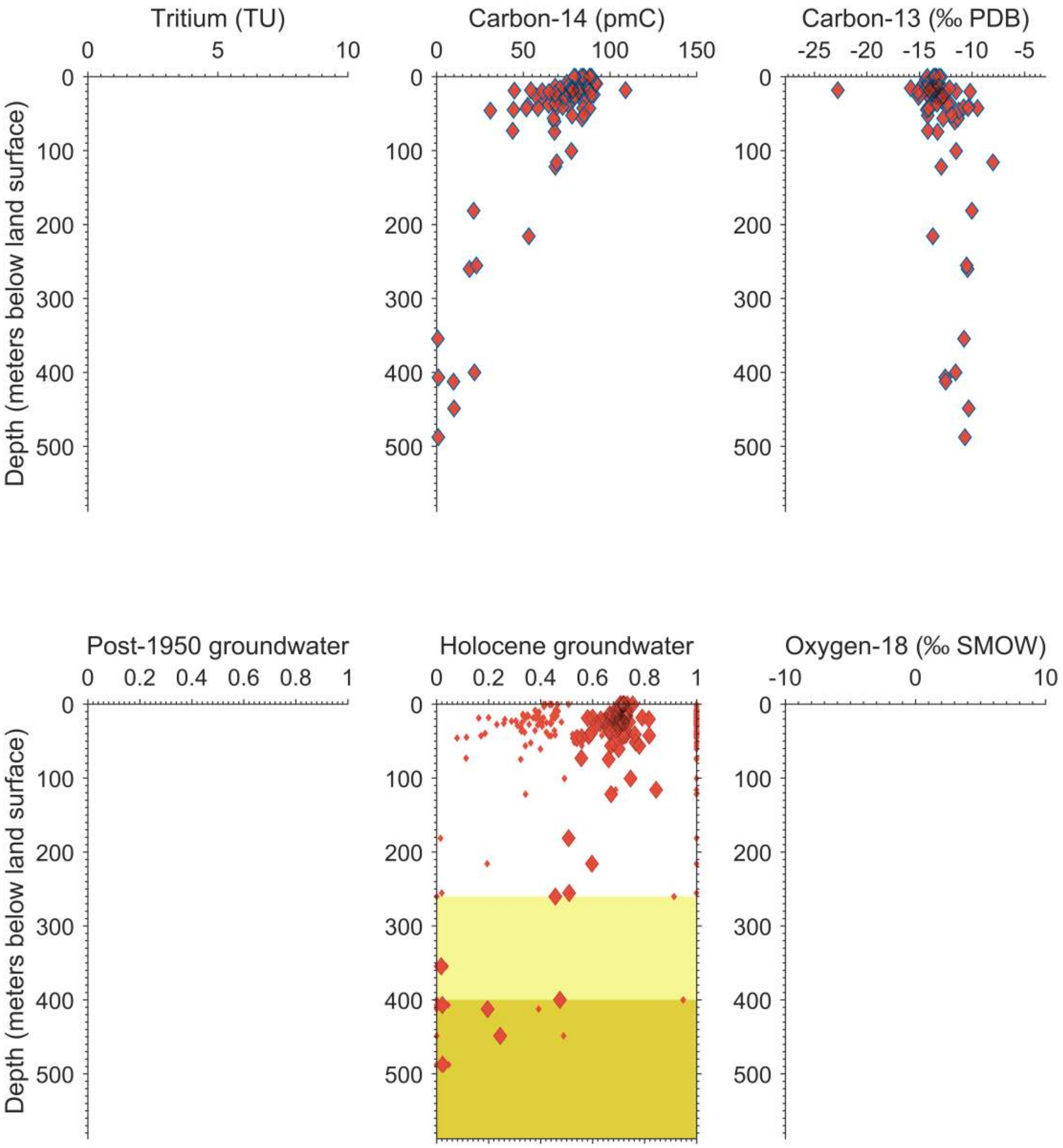
Jacksonville Aquifer, USA



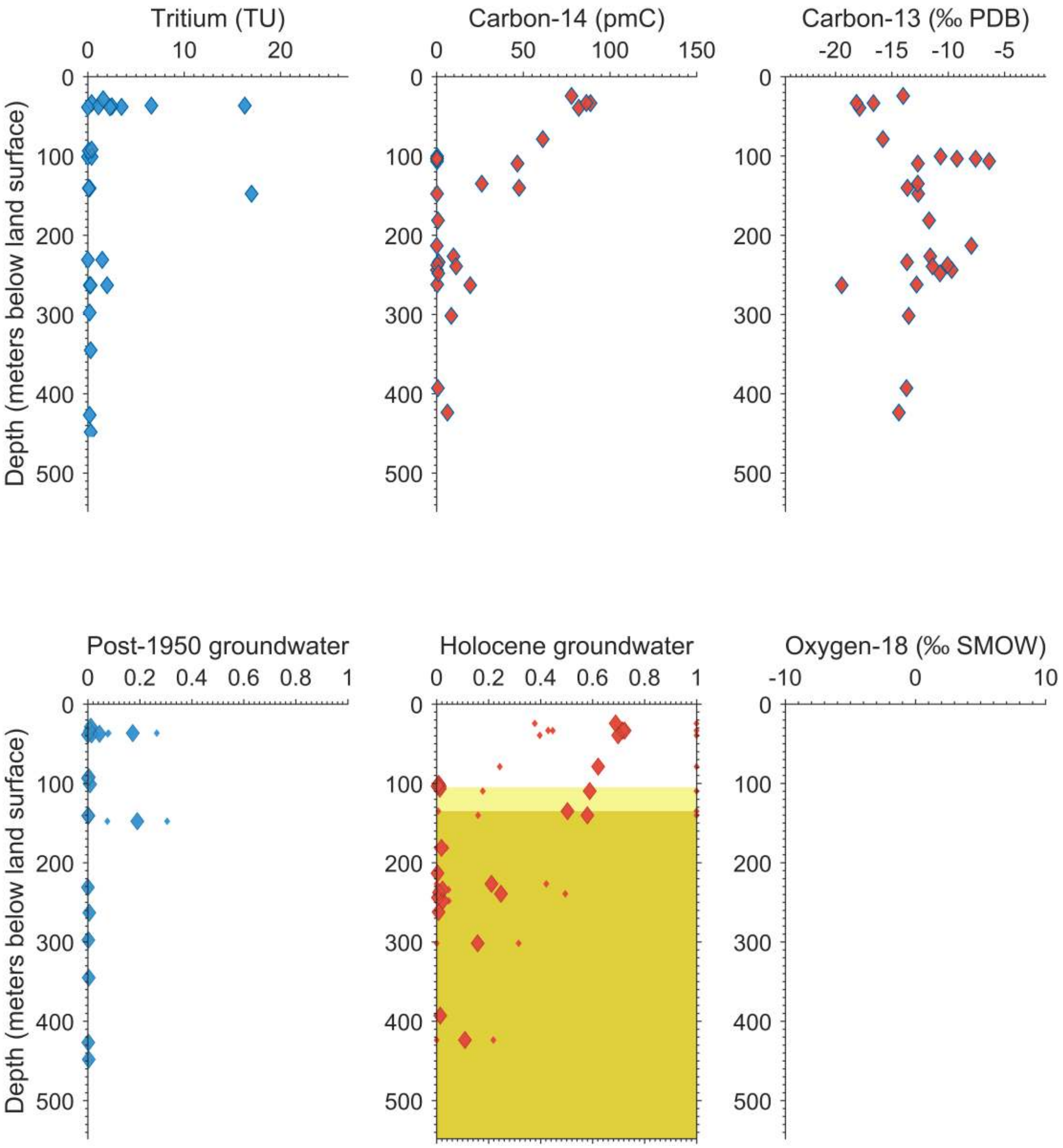
Tampa Bay Aquifer, USA



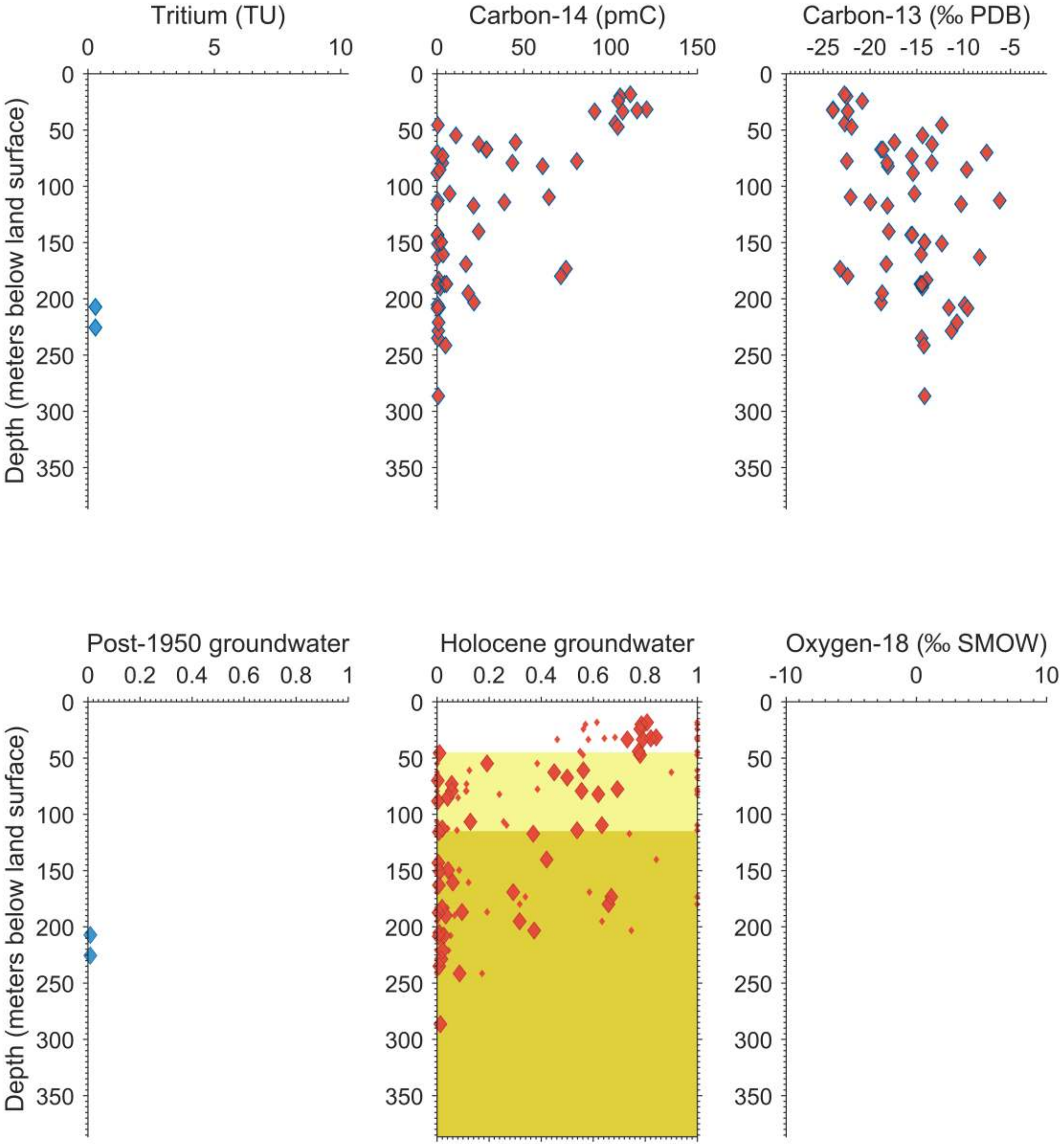
Silurian-Devonian Aquifer, USA



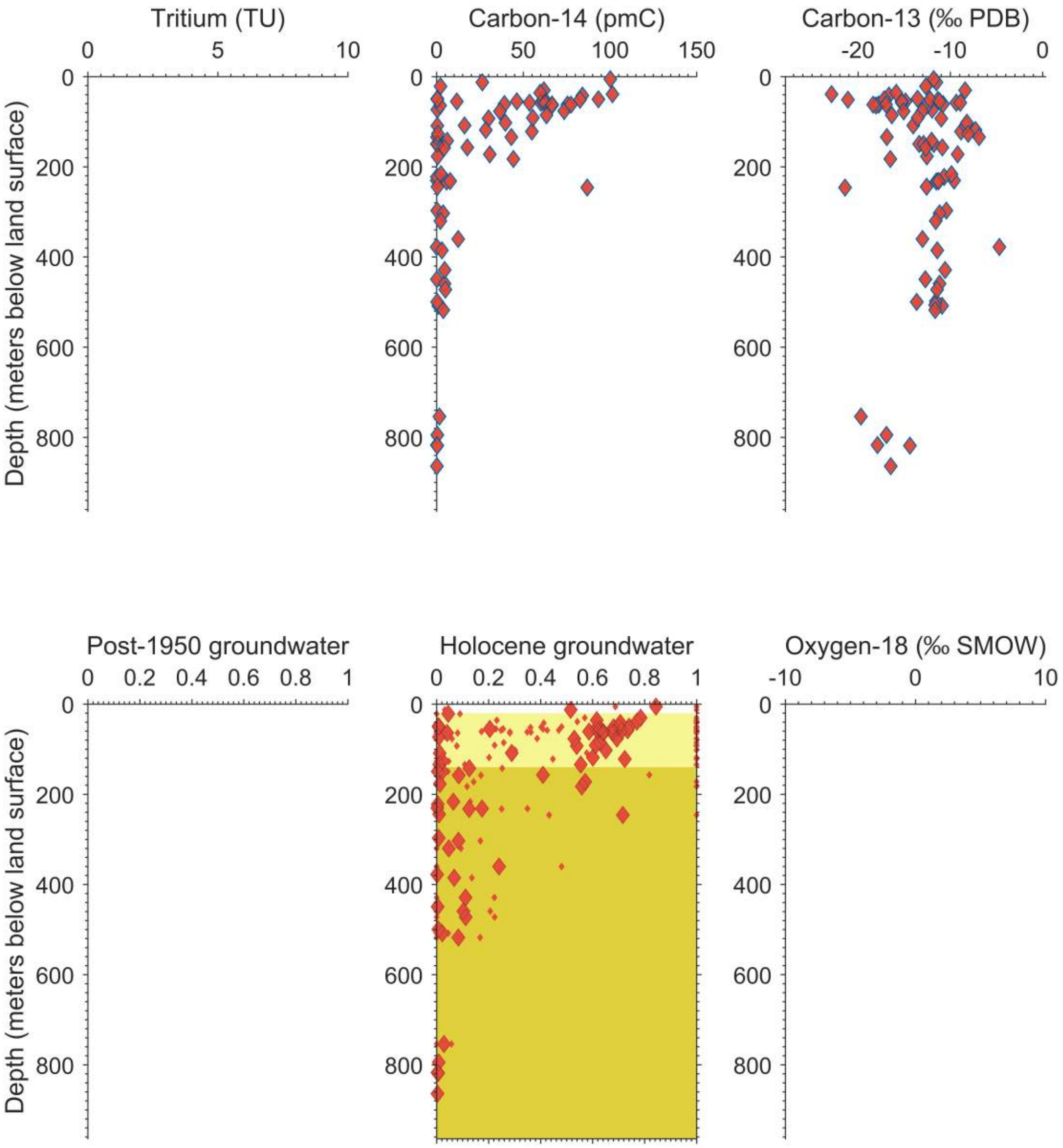
Cambrian Ordovician Sandstone, USA



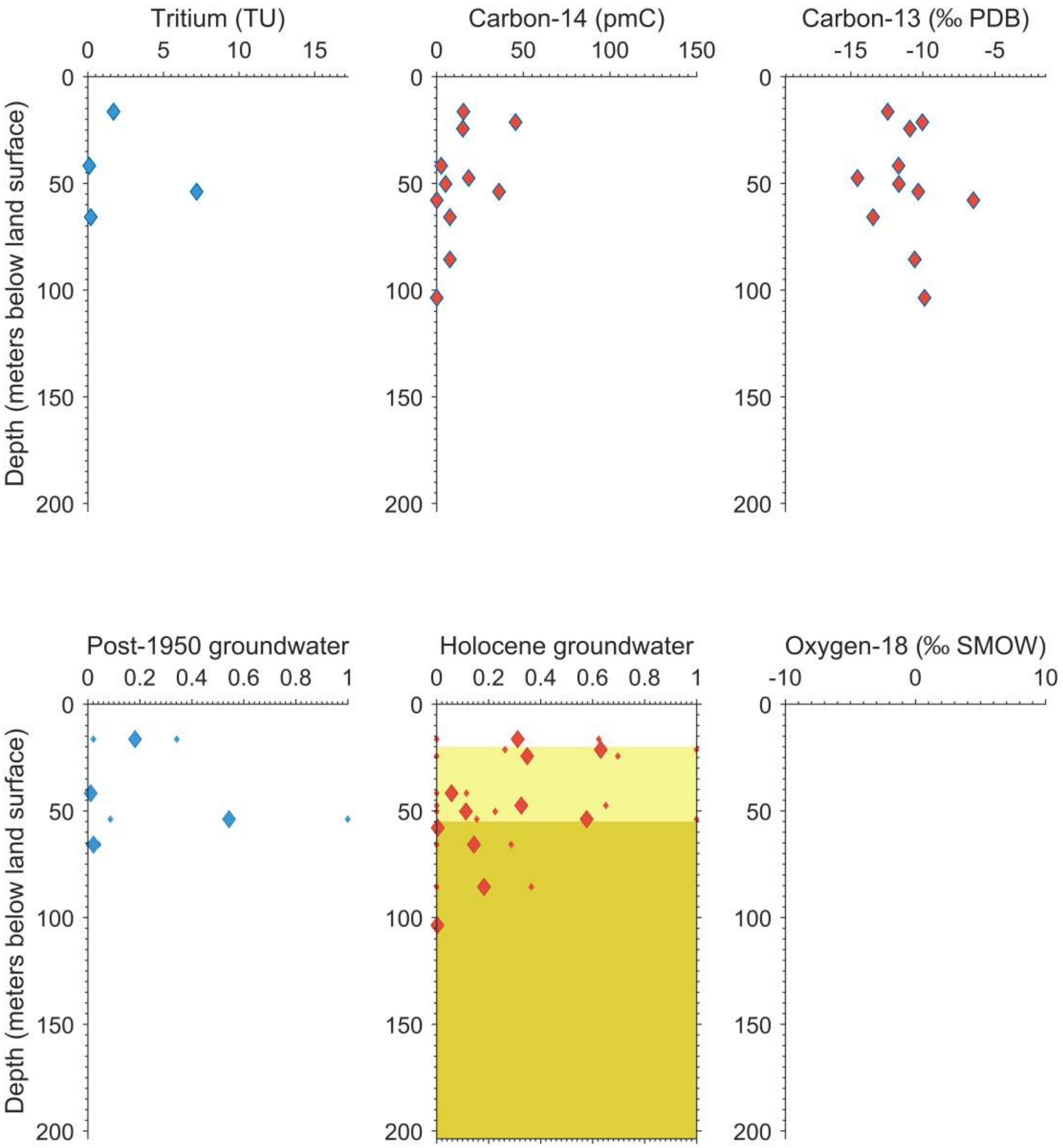
Mississippi Alluvial Aquifer, USA



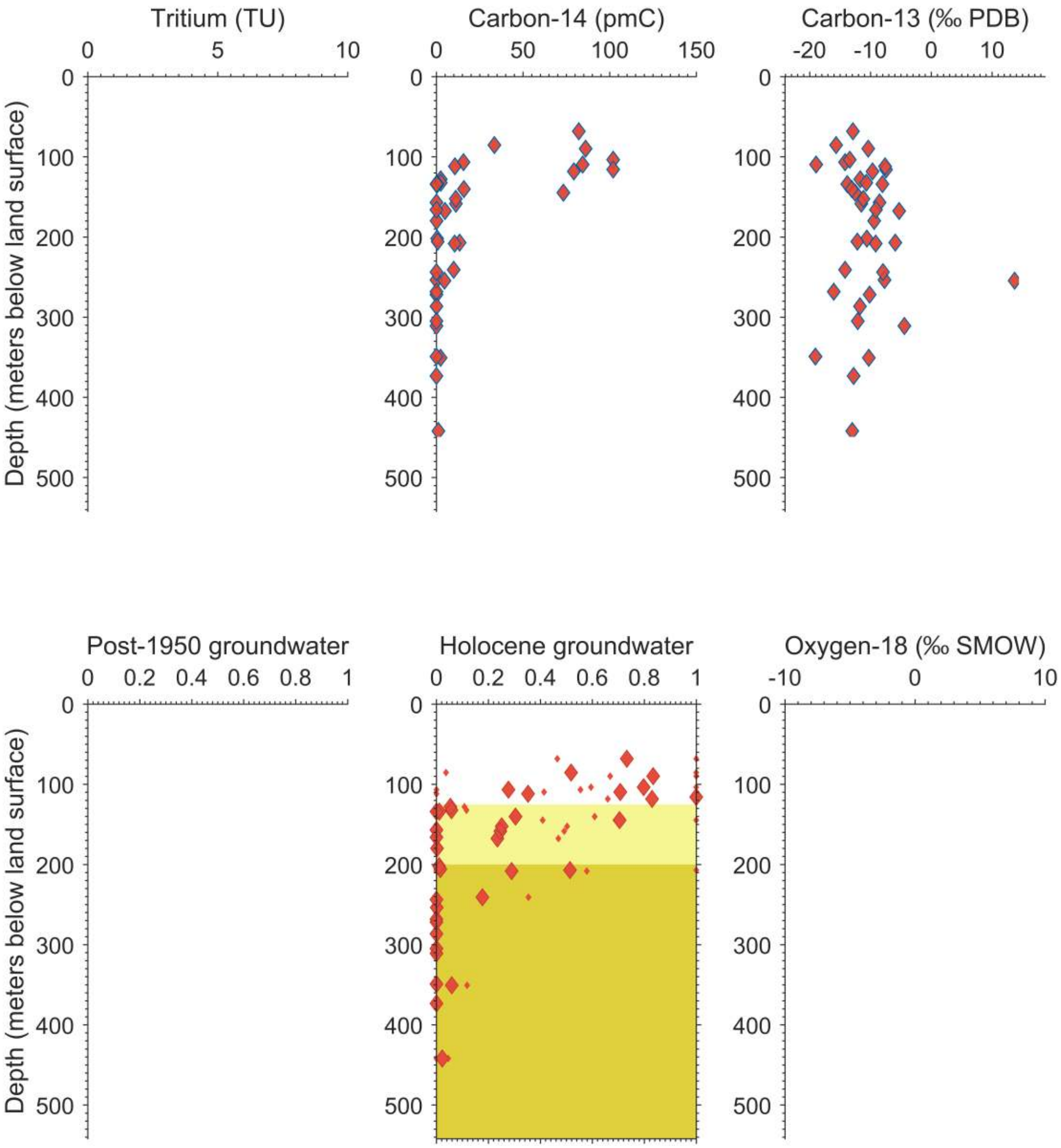
Mississippi Embayment, USA



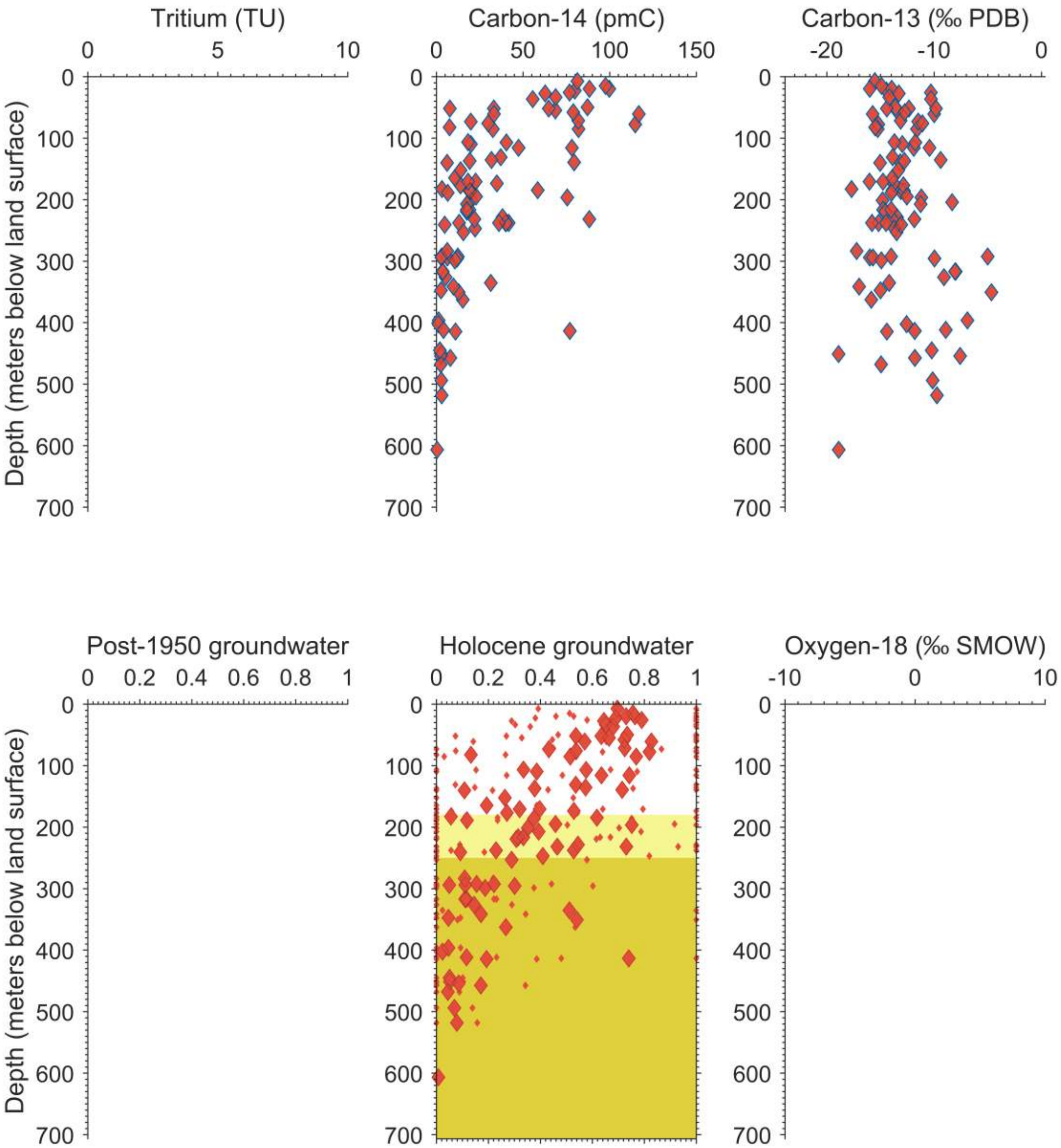
Houston Coastal Lowlands, USA



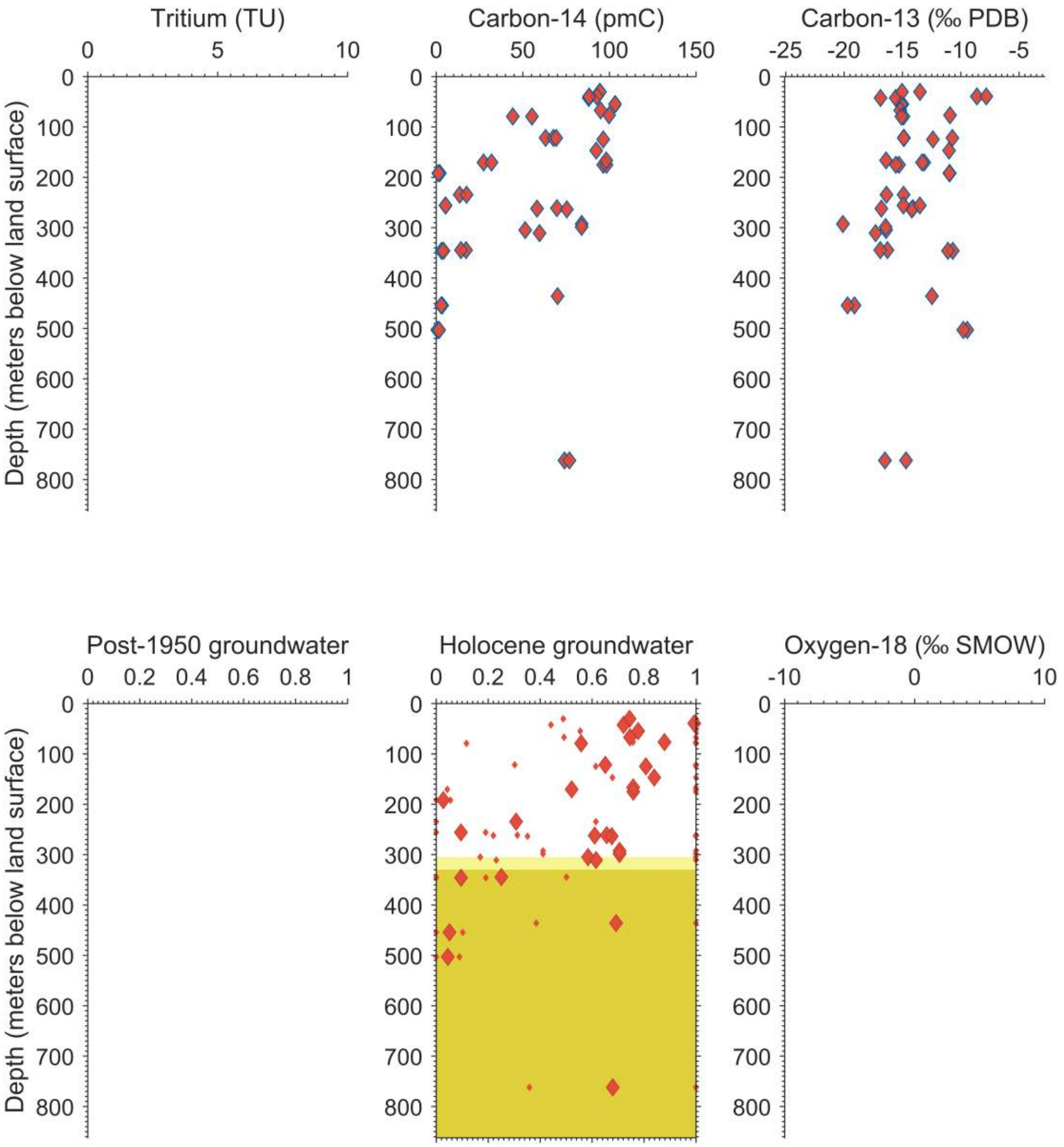
Lower Tertiary Aquifers, USA



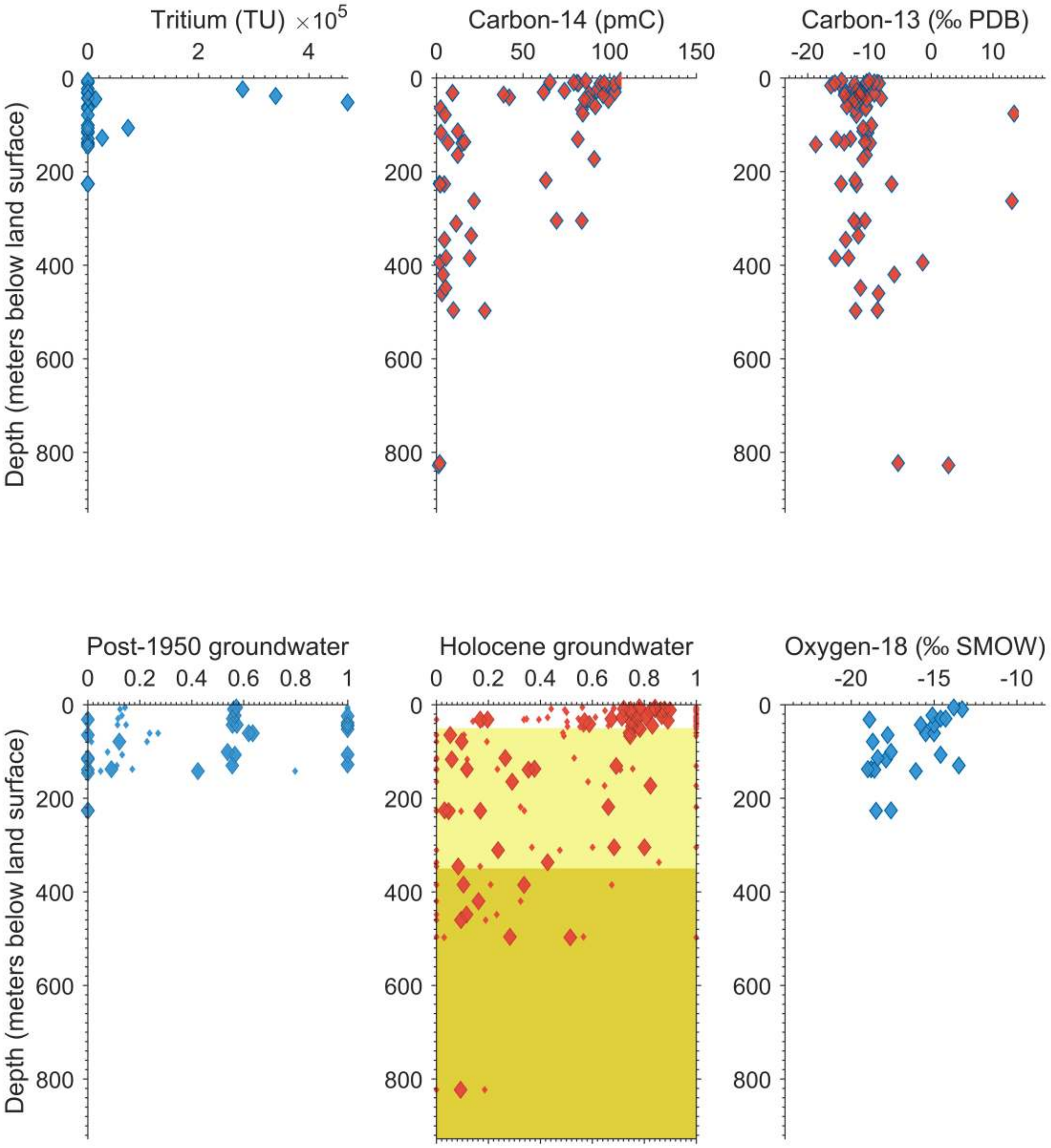
Denver Basin, USA



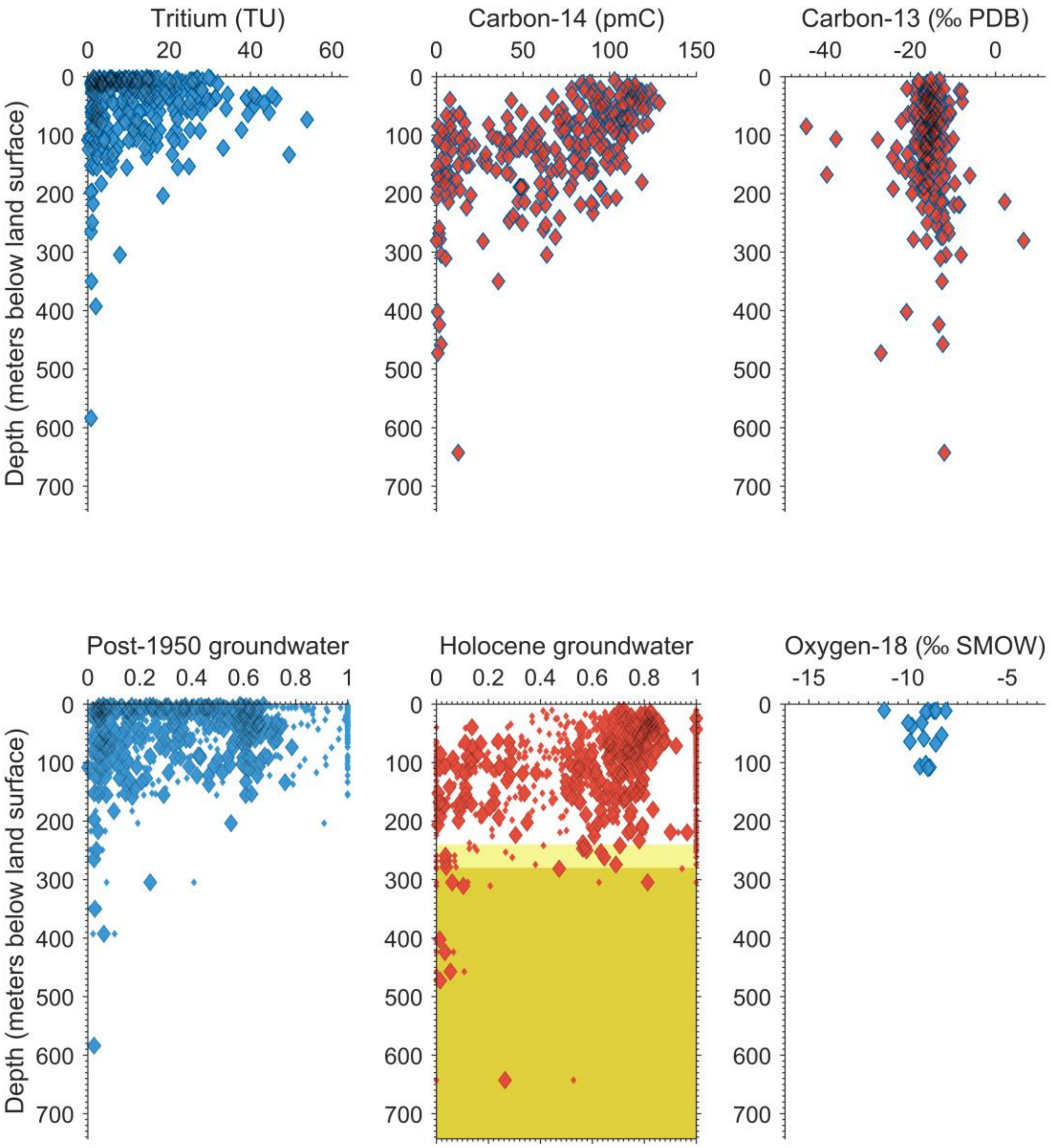
San Diego Basin, USA



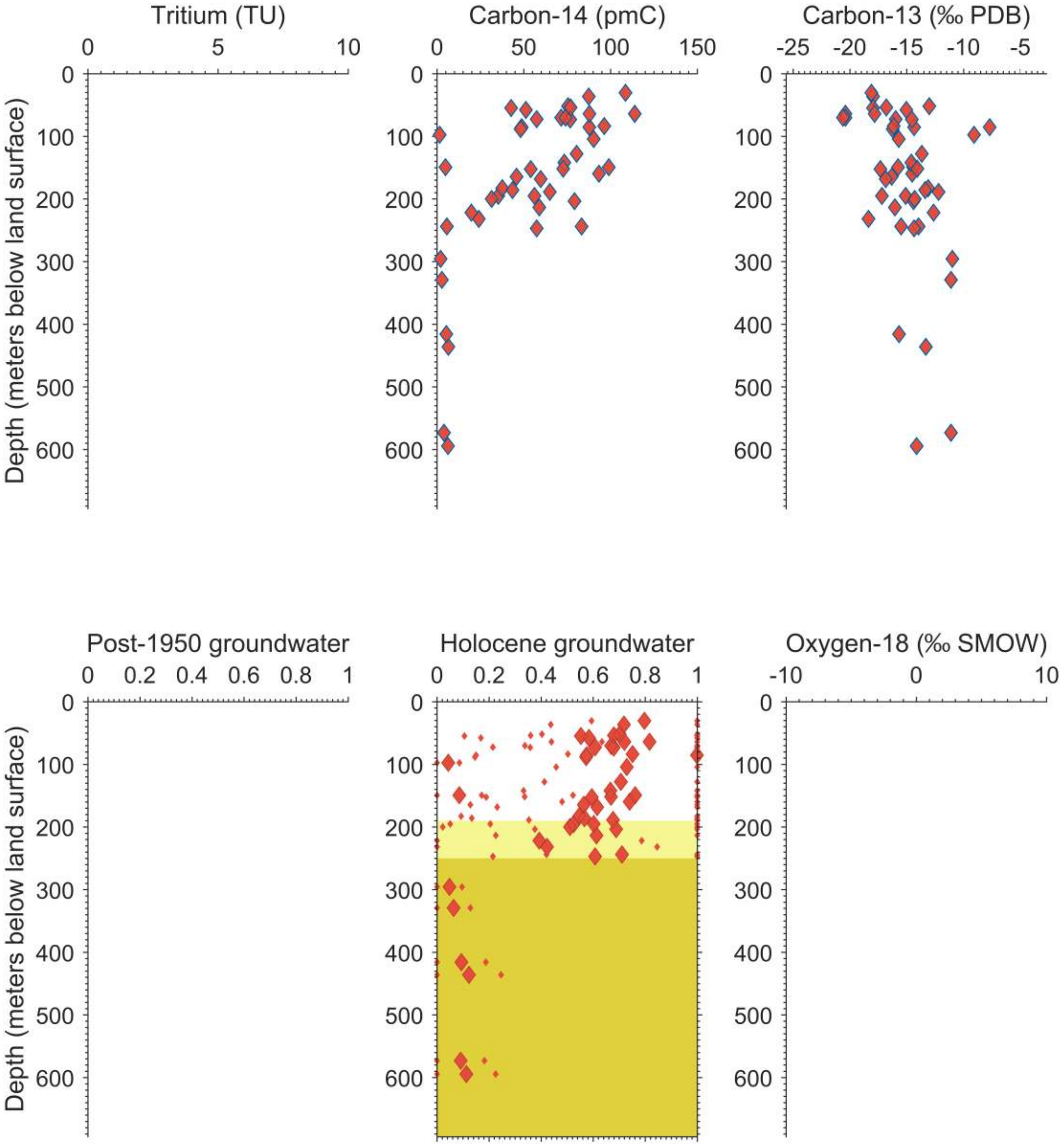
Temecula-Murrieta Basin, USA



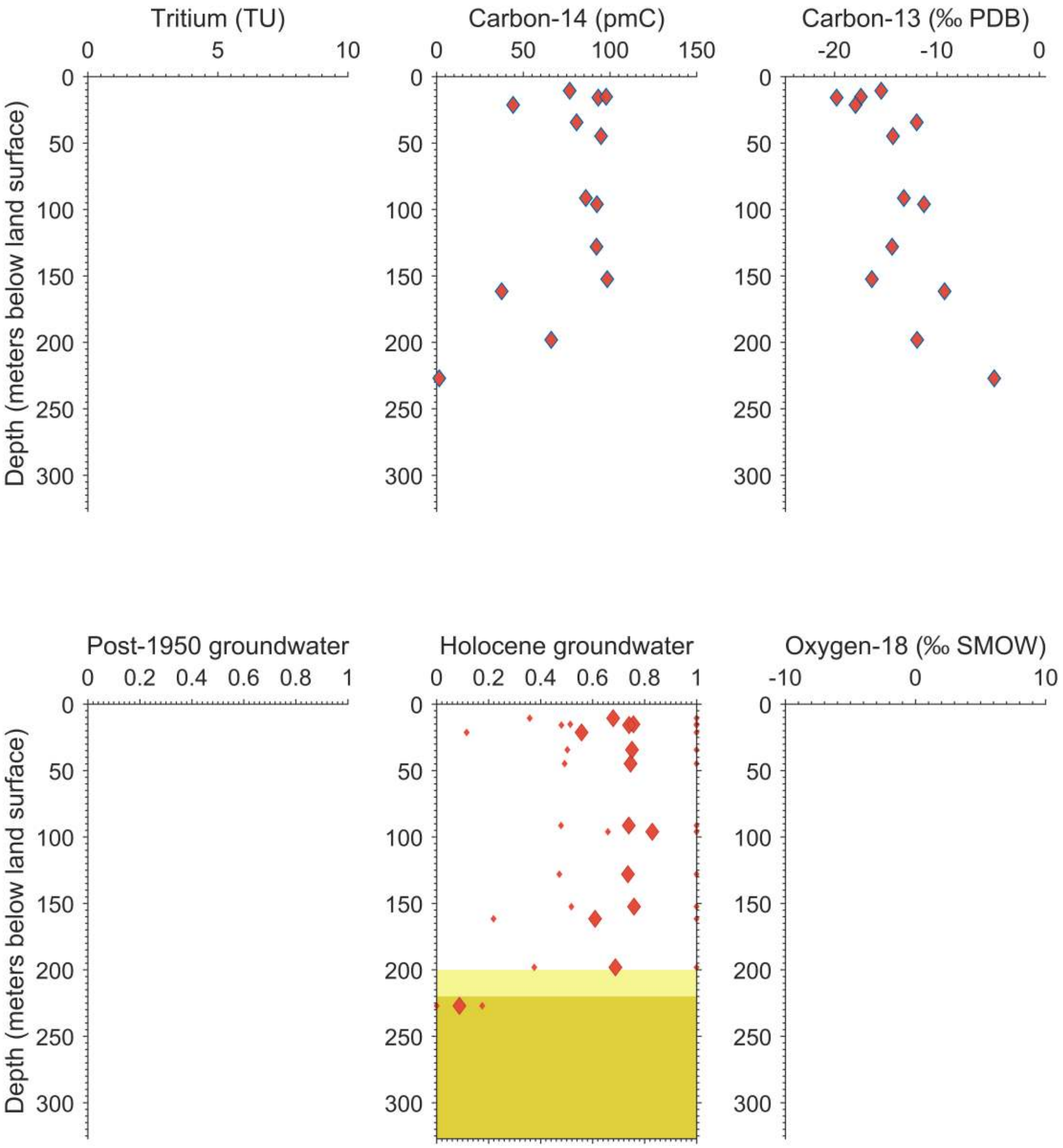
Columbia Basin, USA



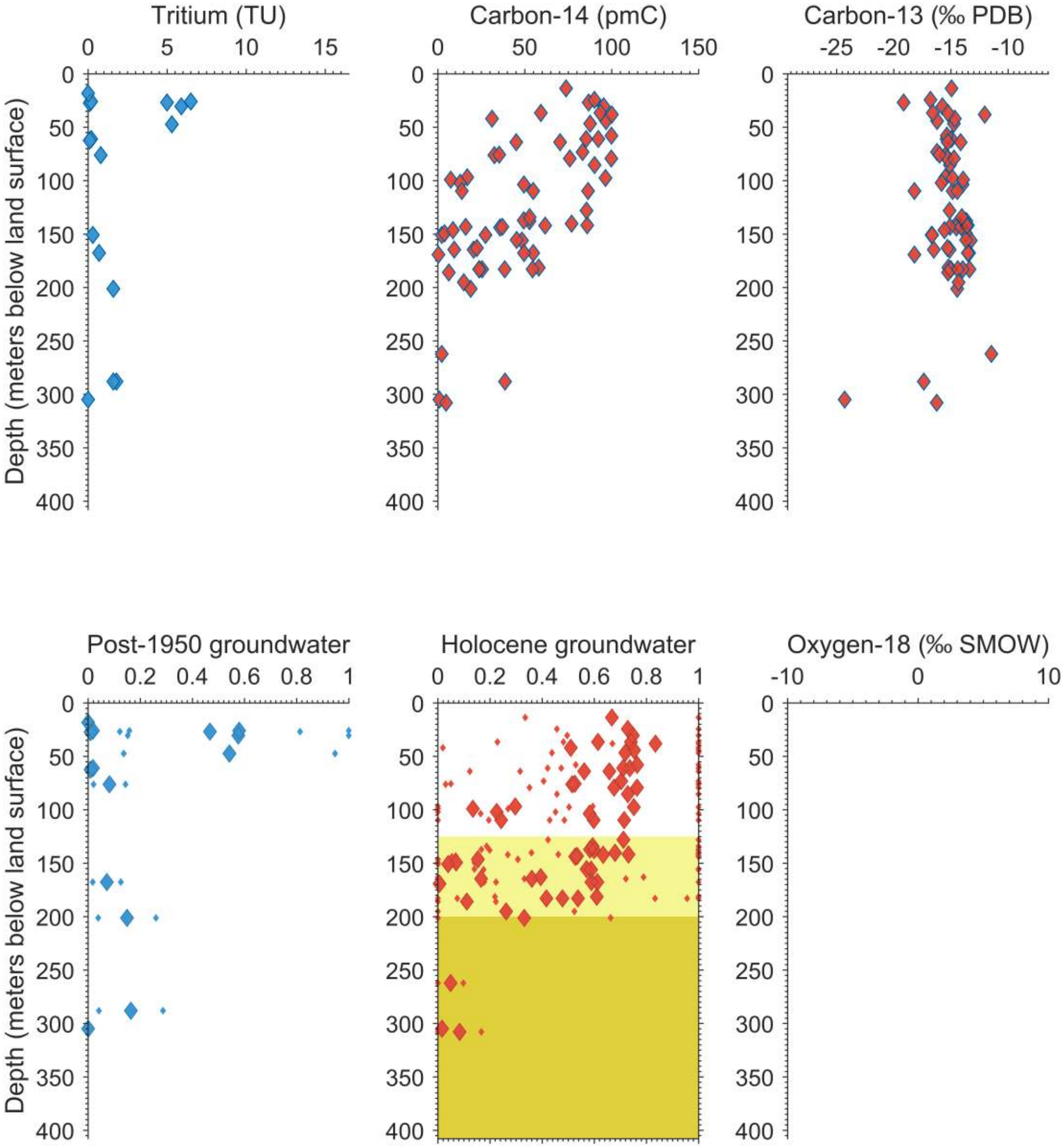
Southern Central Valley, USA



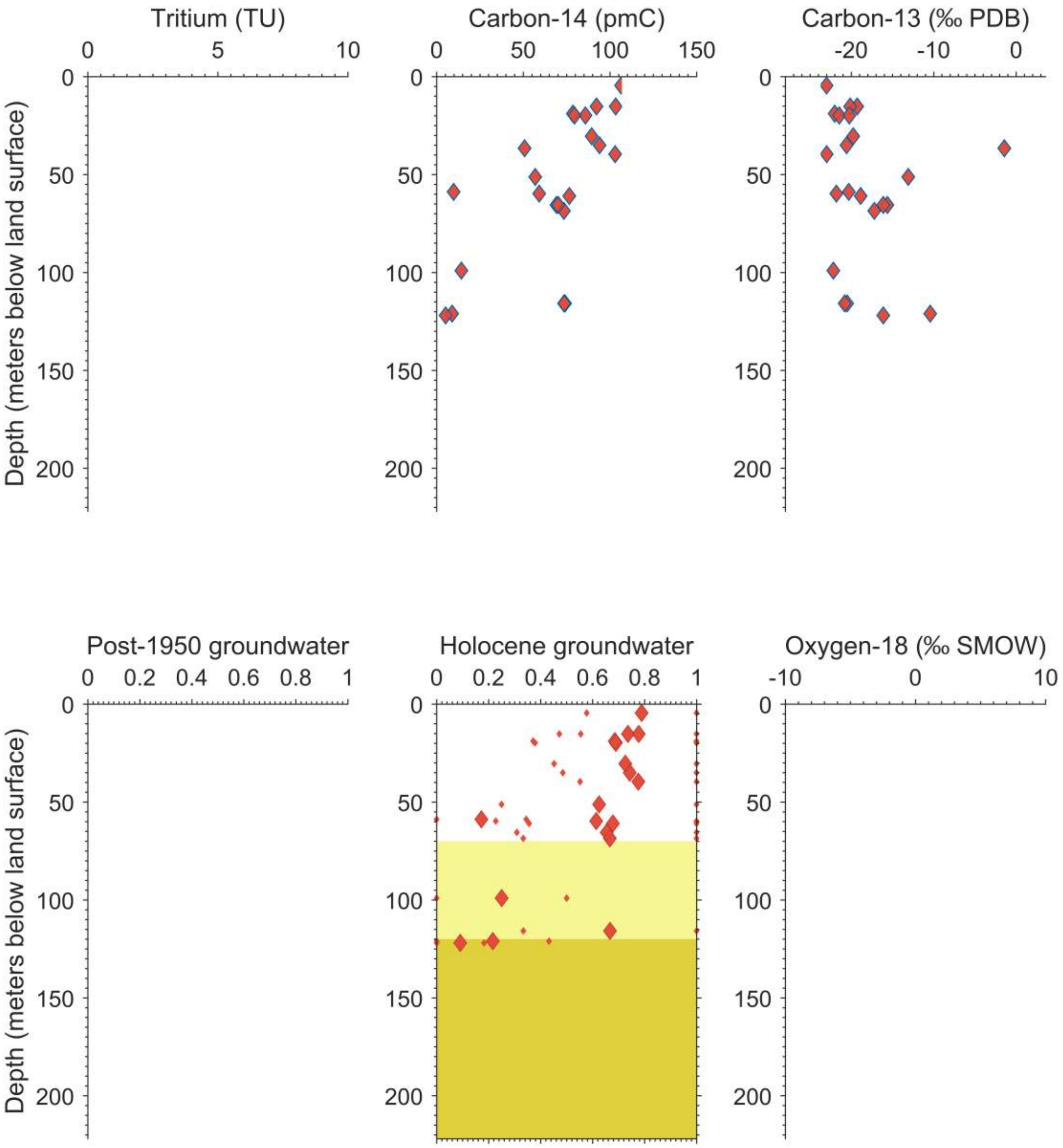
California Bay Area: Monterey Bay, USA



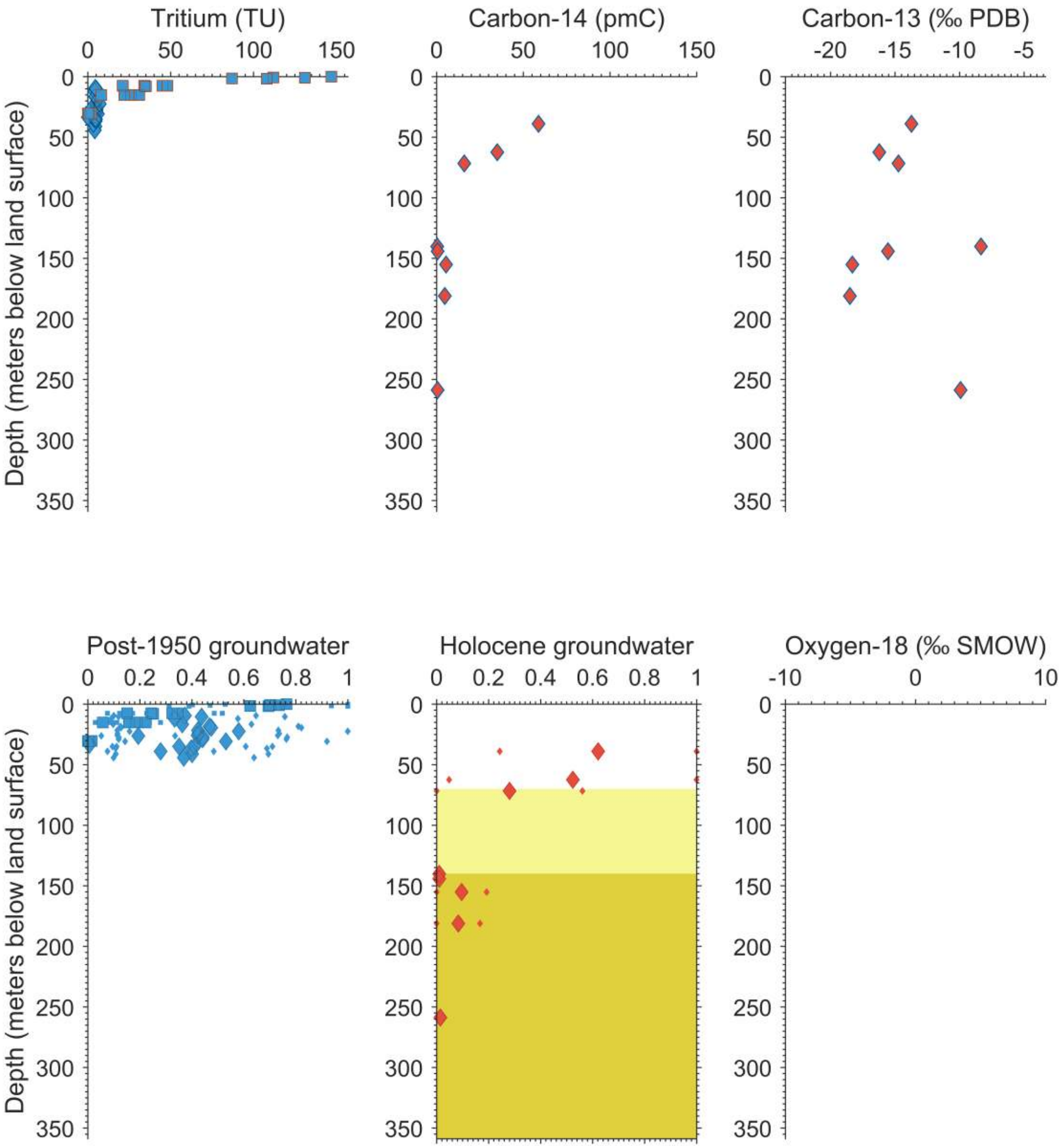
Livermore Valley, USA



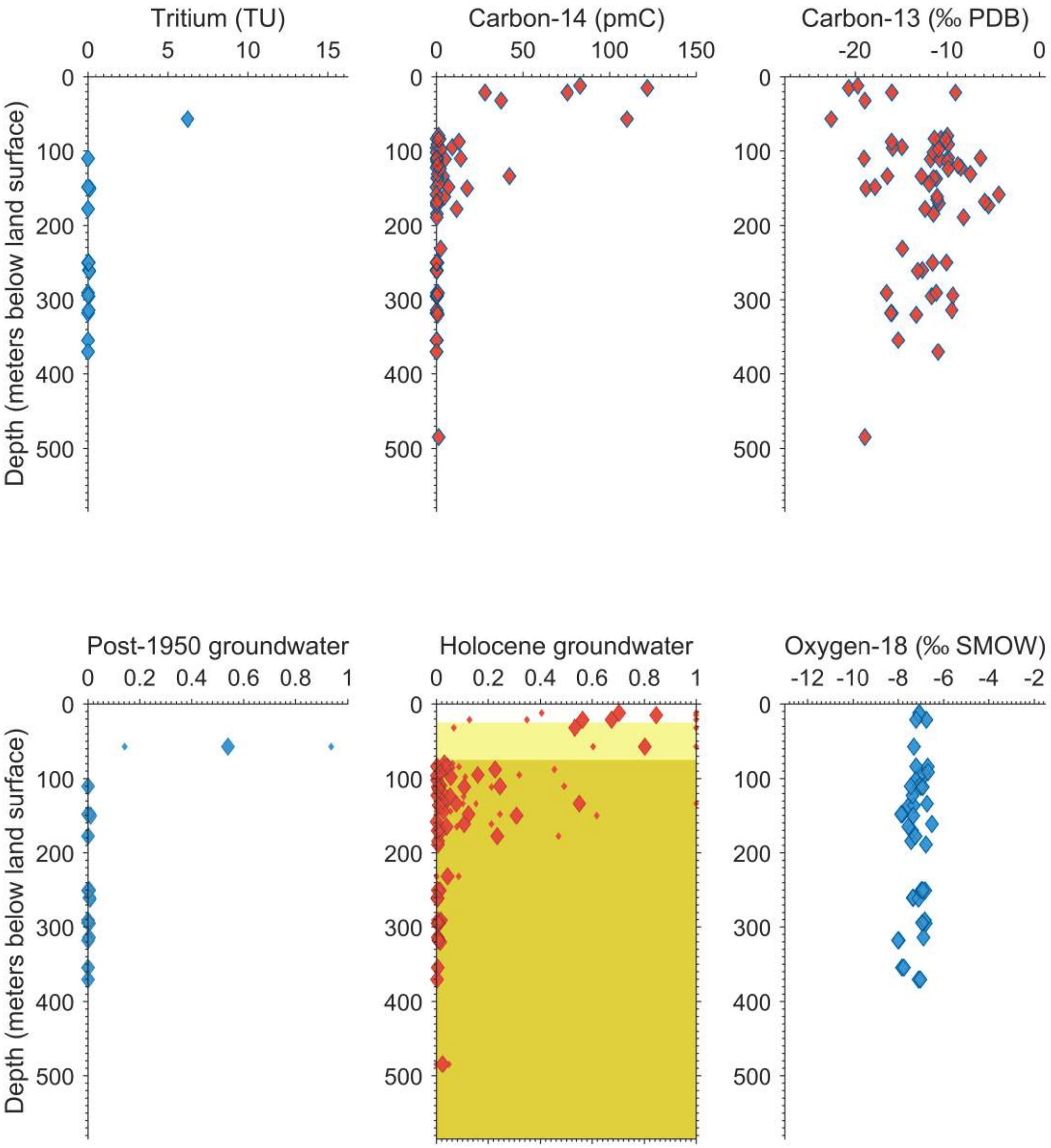
California Bay Area: East, USA



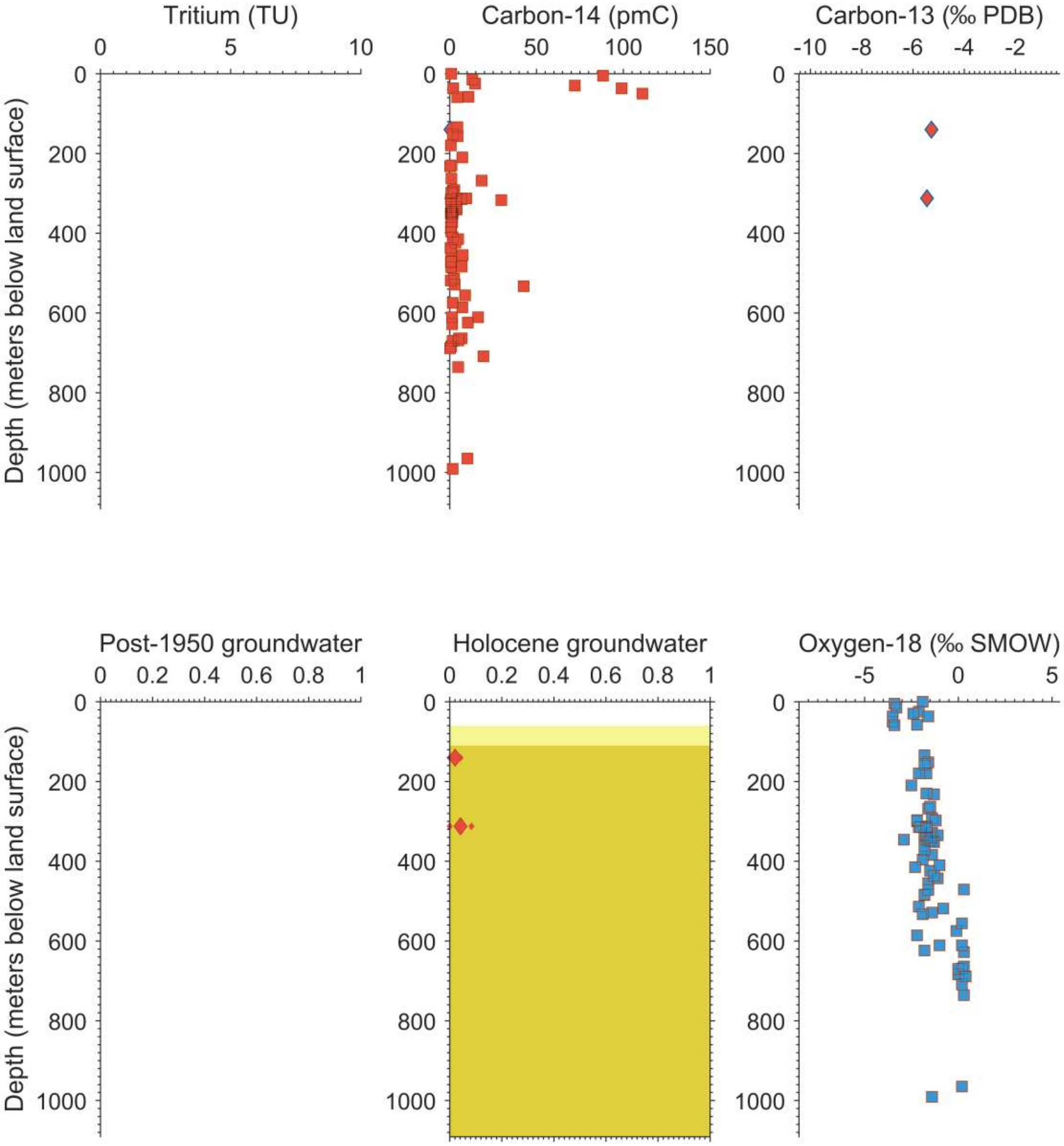
Eureka Coast, USA



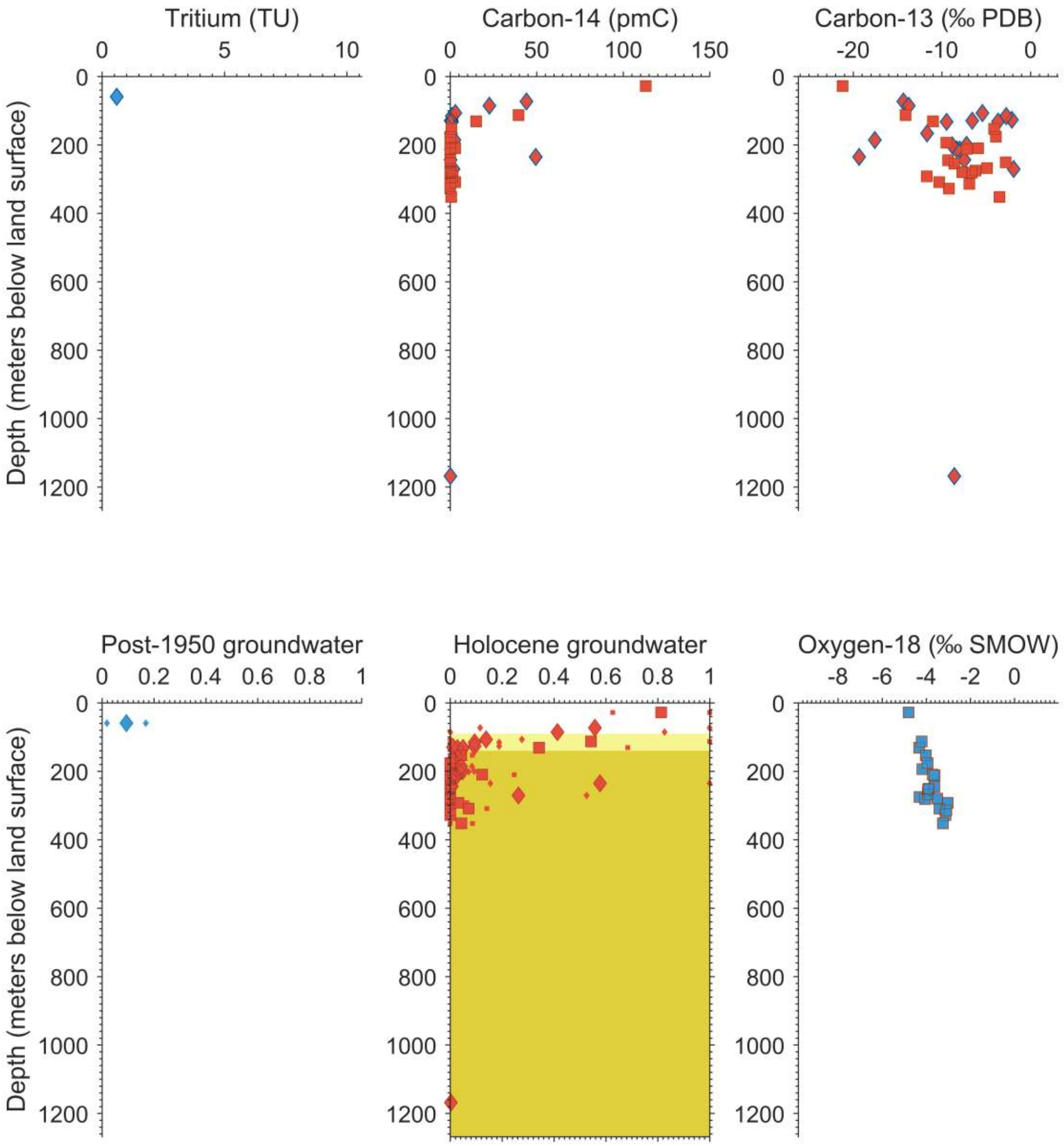
Wharton tract, USA



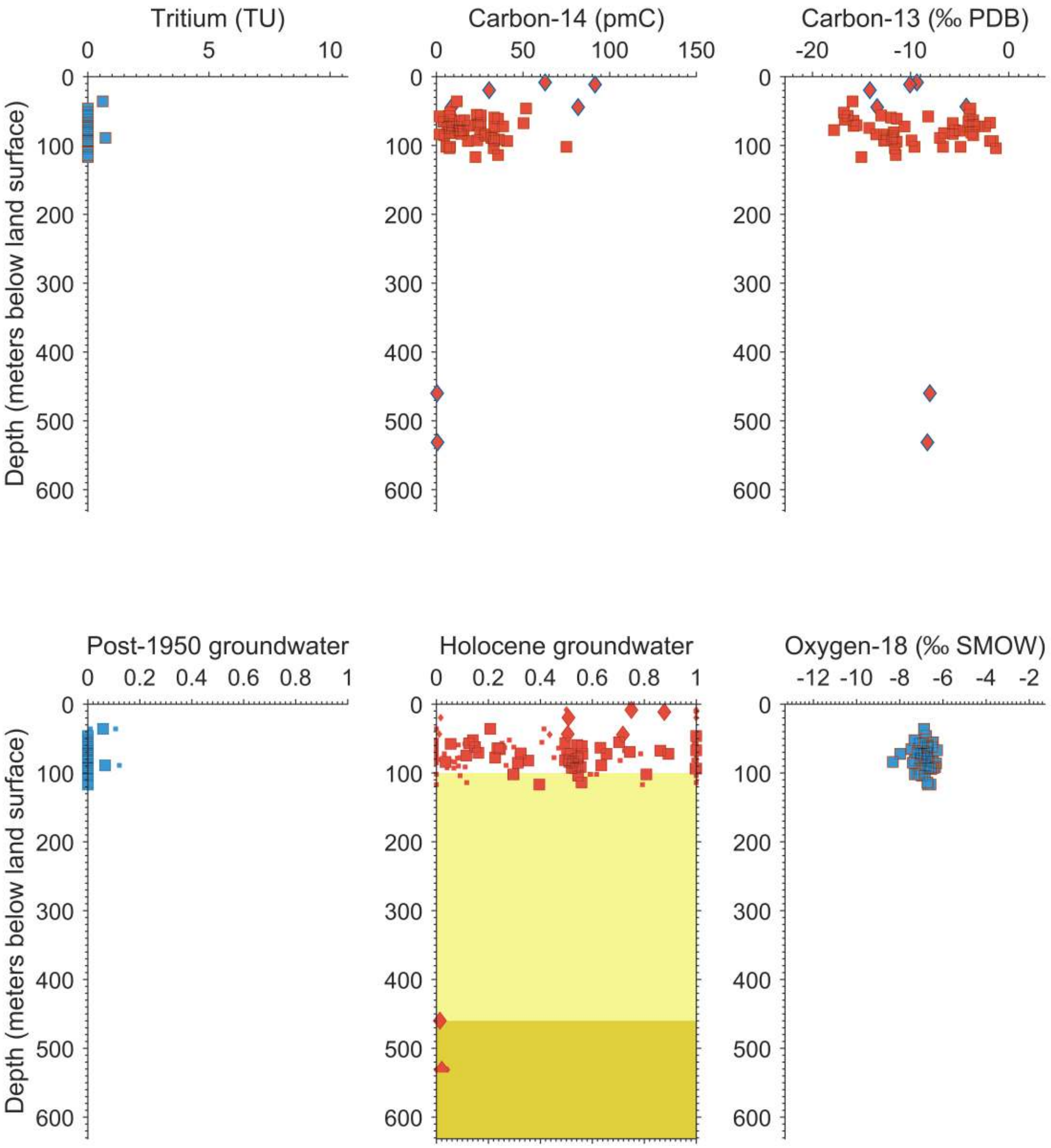
Maryland Coastal Plain (Aquia, Patapsco), USA



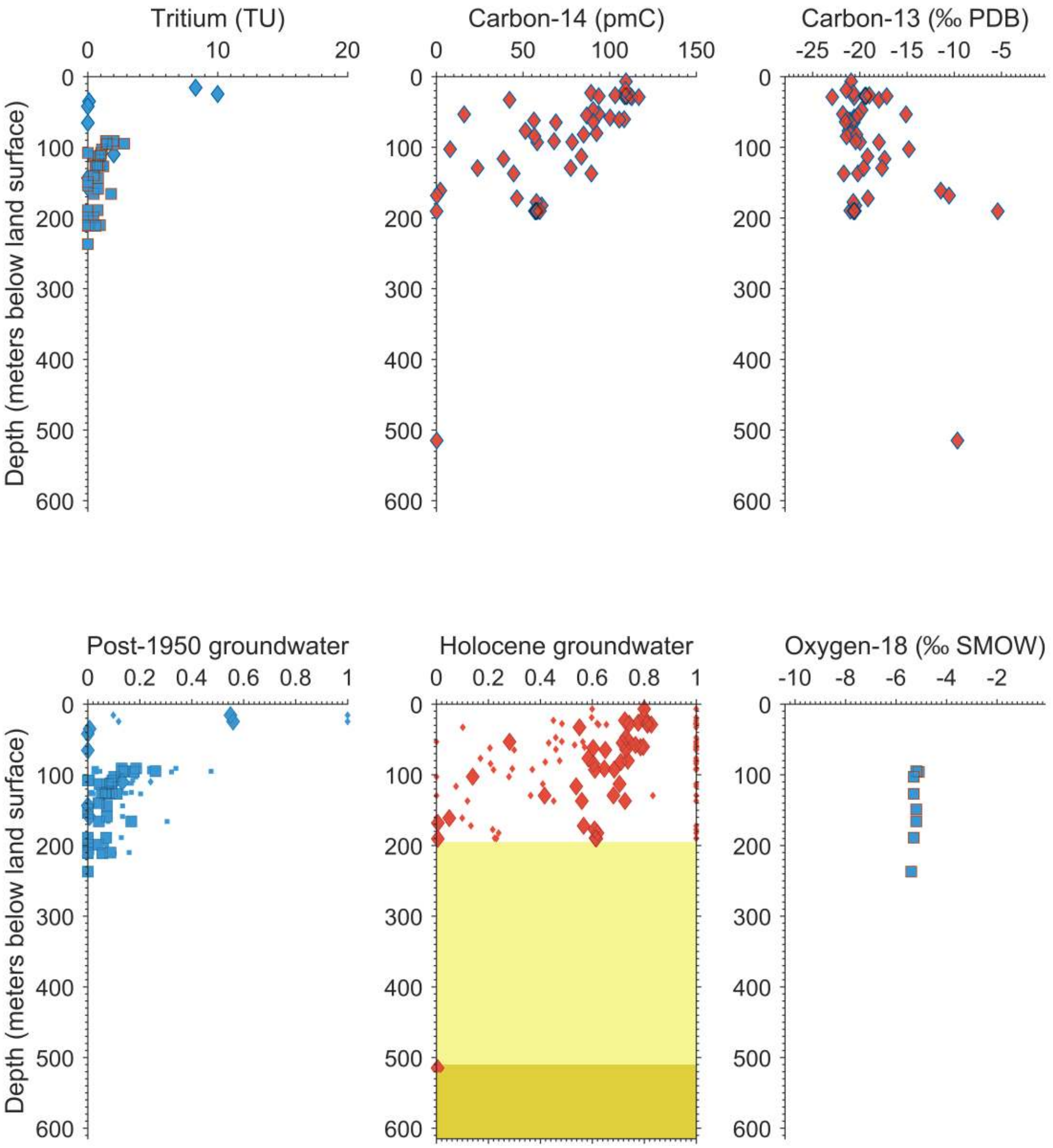
Floridan Surficial Aquifers, USA (Paleowater transition based on oxygen-18)



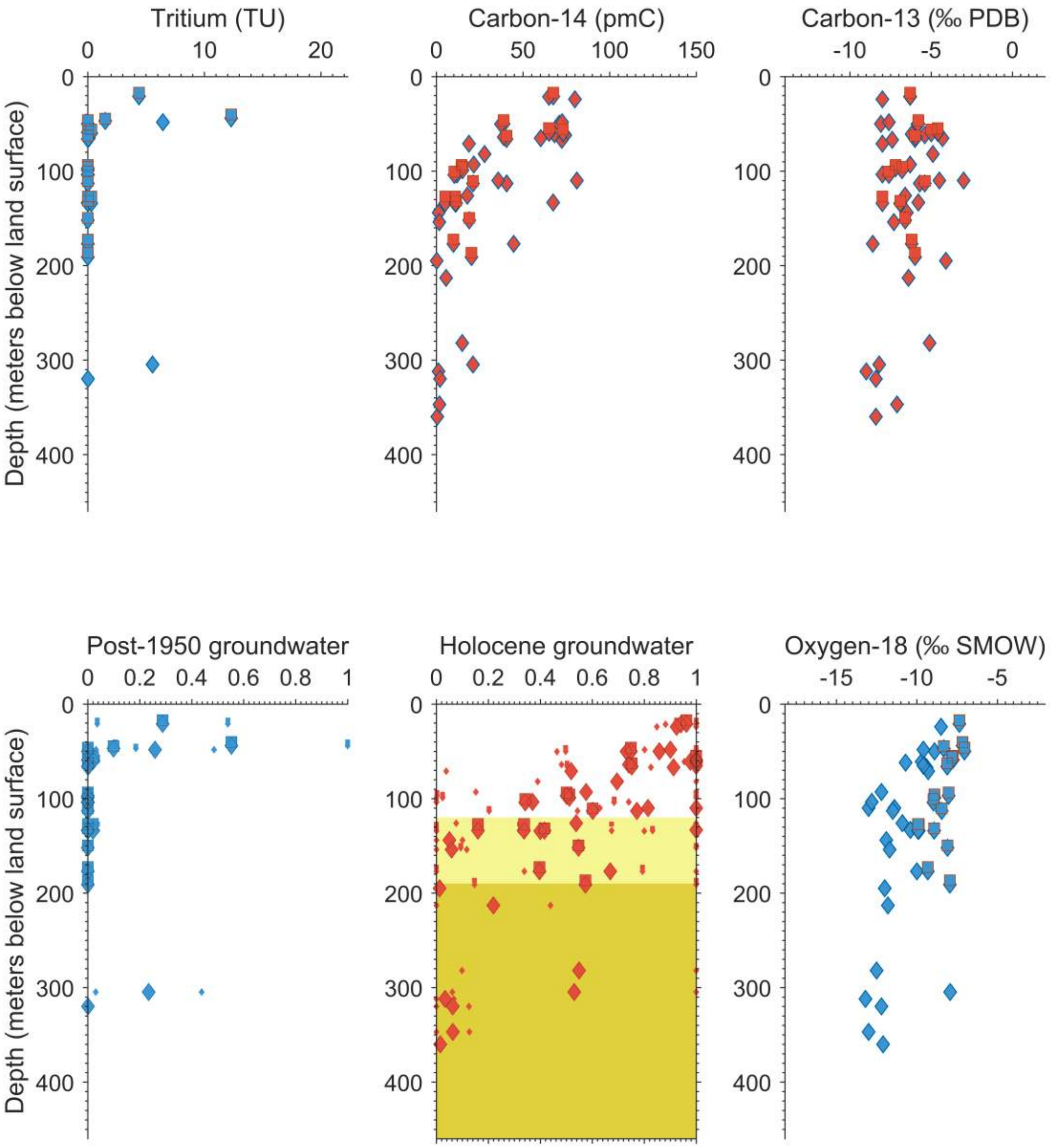
Floridan Aquifer System, USA



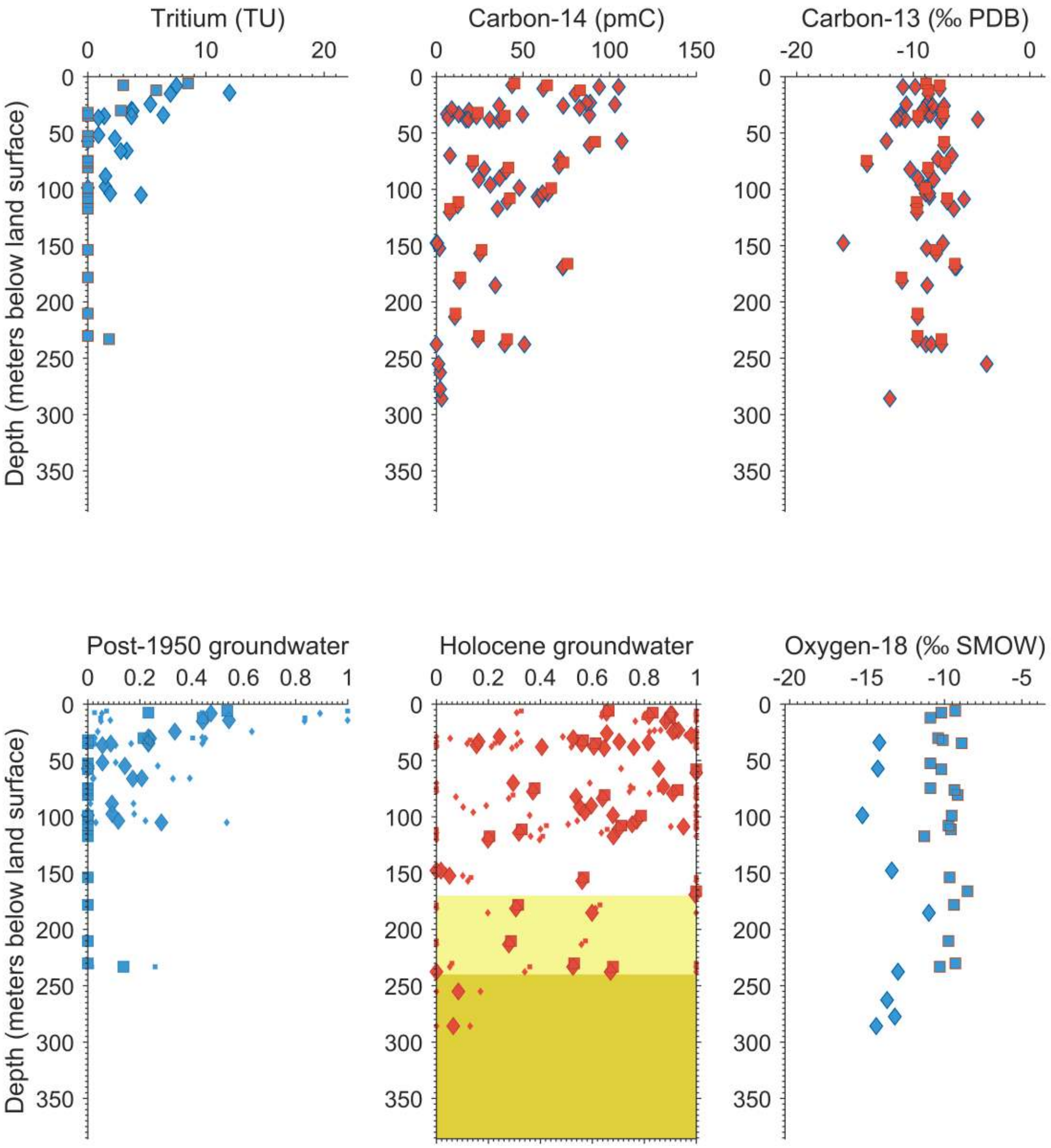
Mahomet Aquifer, USA



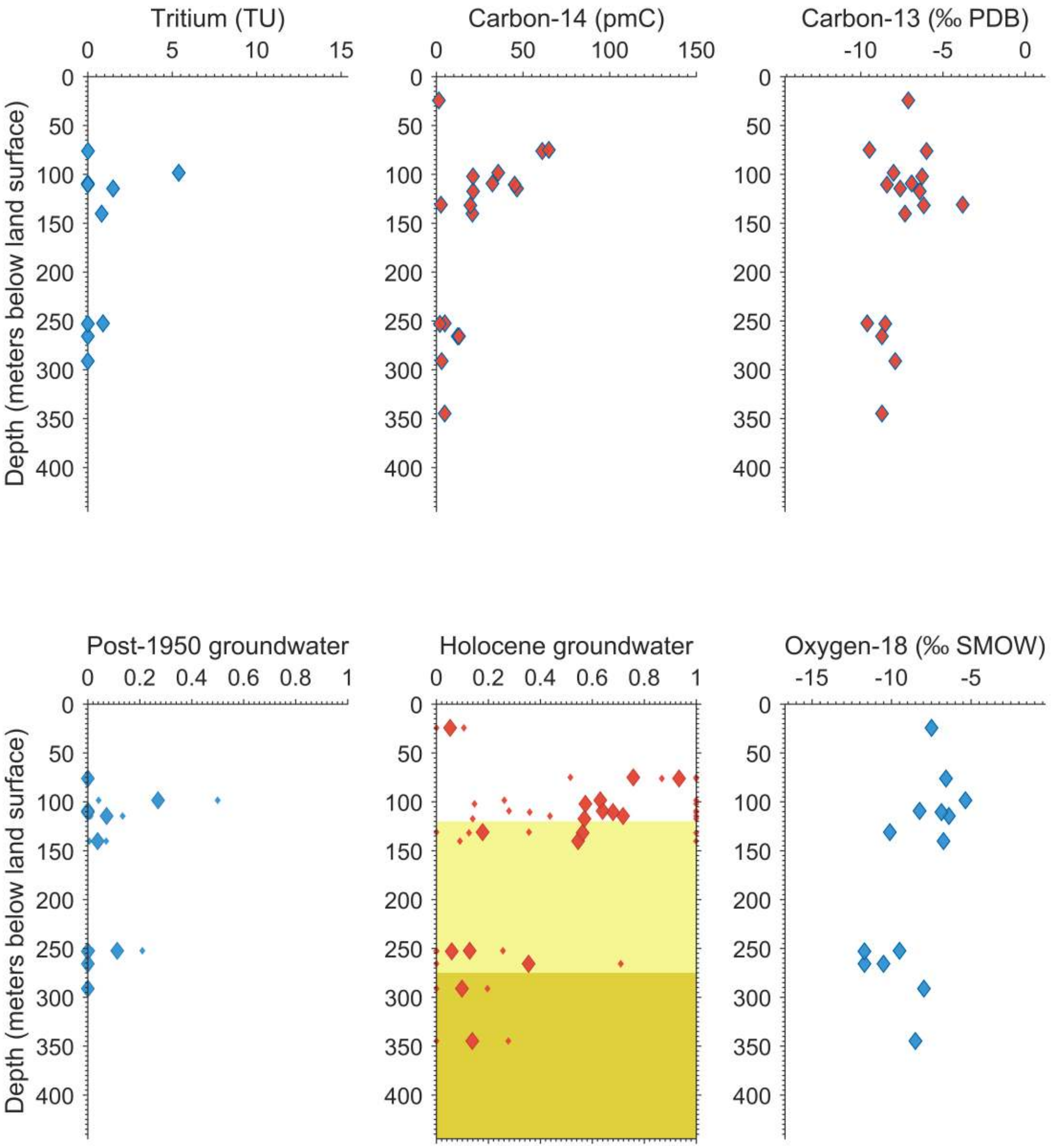
Mississippi embayment: Memphis aquifer, USA



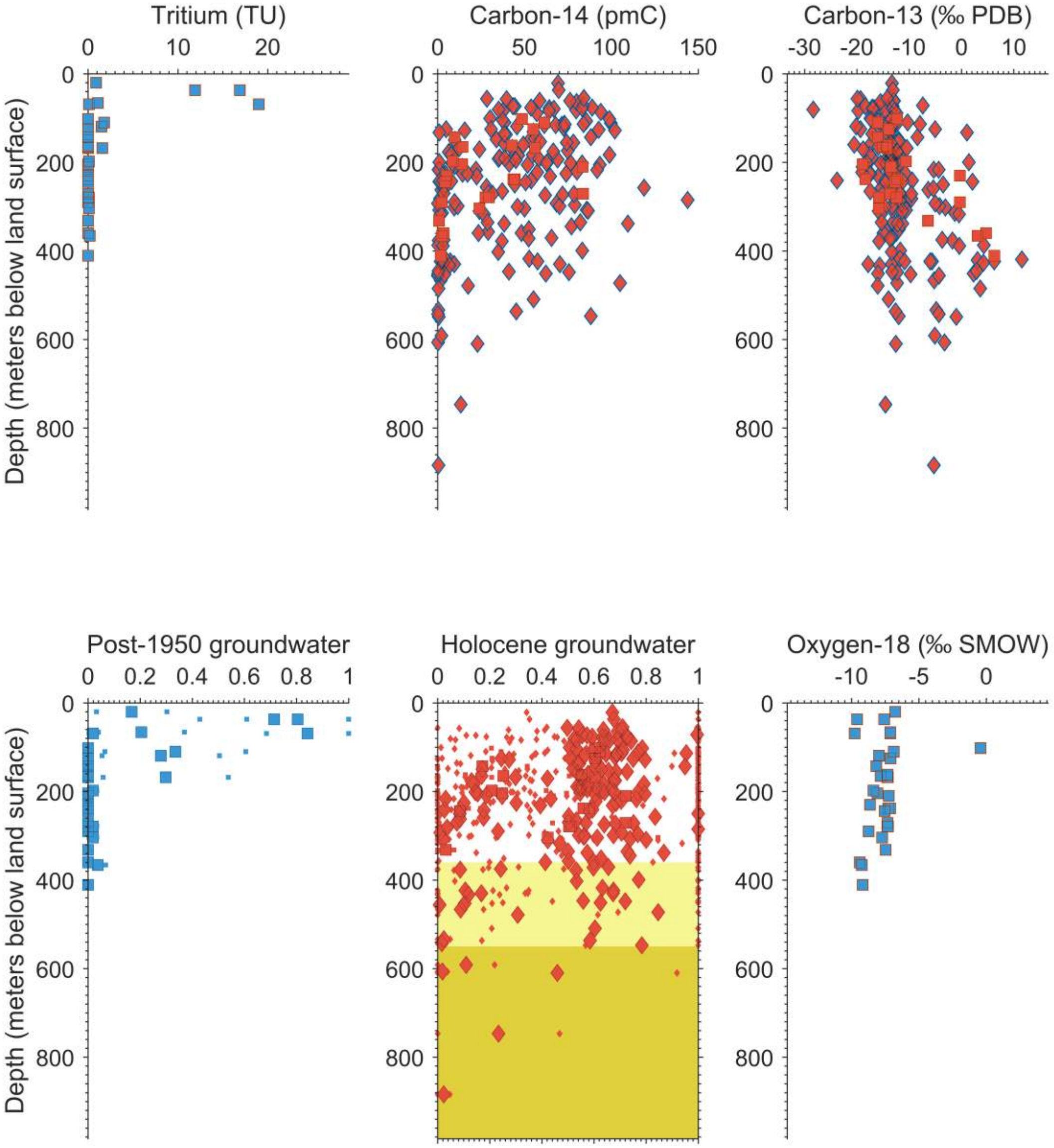
High Plains Aquifer: Central (Ogallala), USA



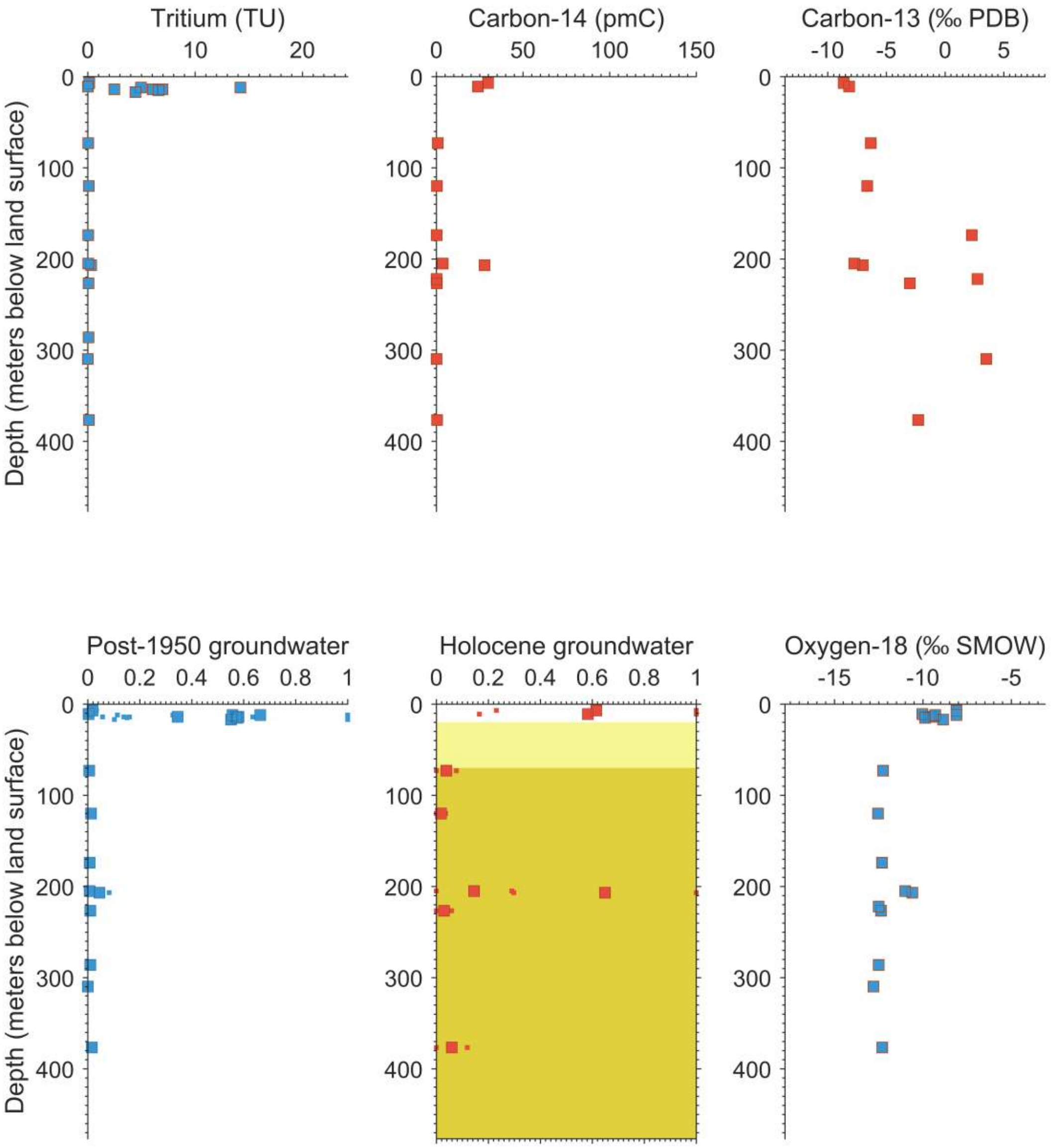
High Plains Aquifer: North (Ogallala), USA



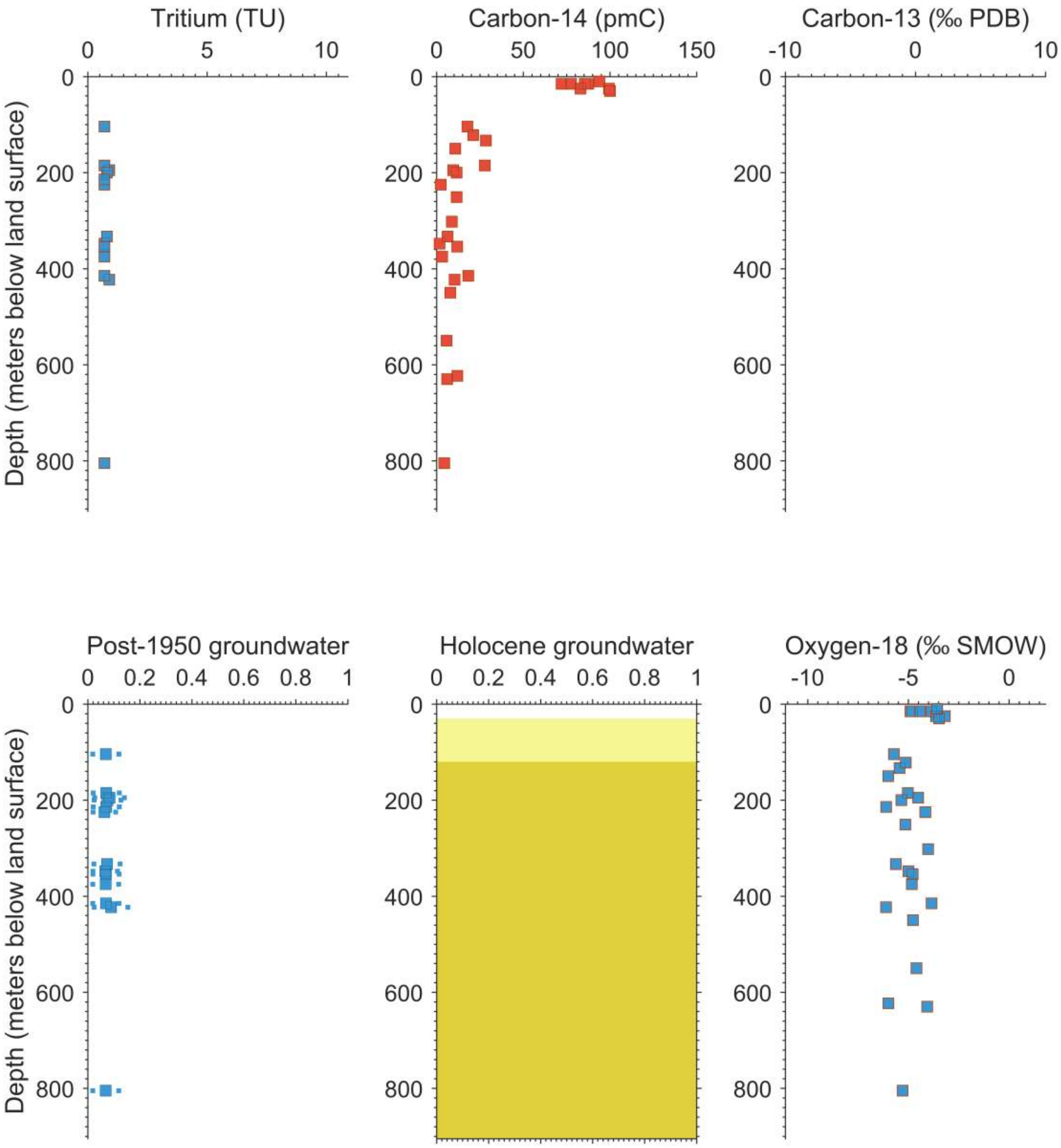
High Plains Aquifer: South (Dockum), USA



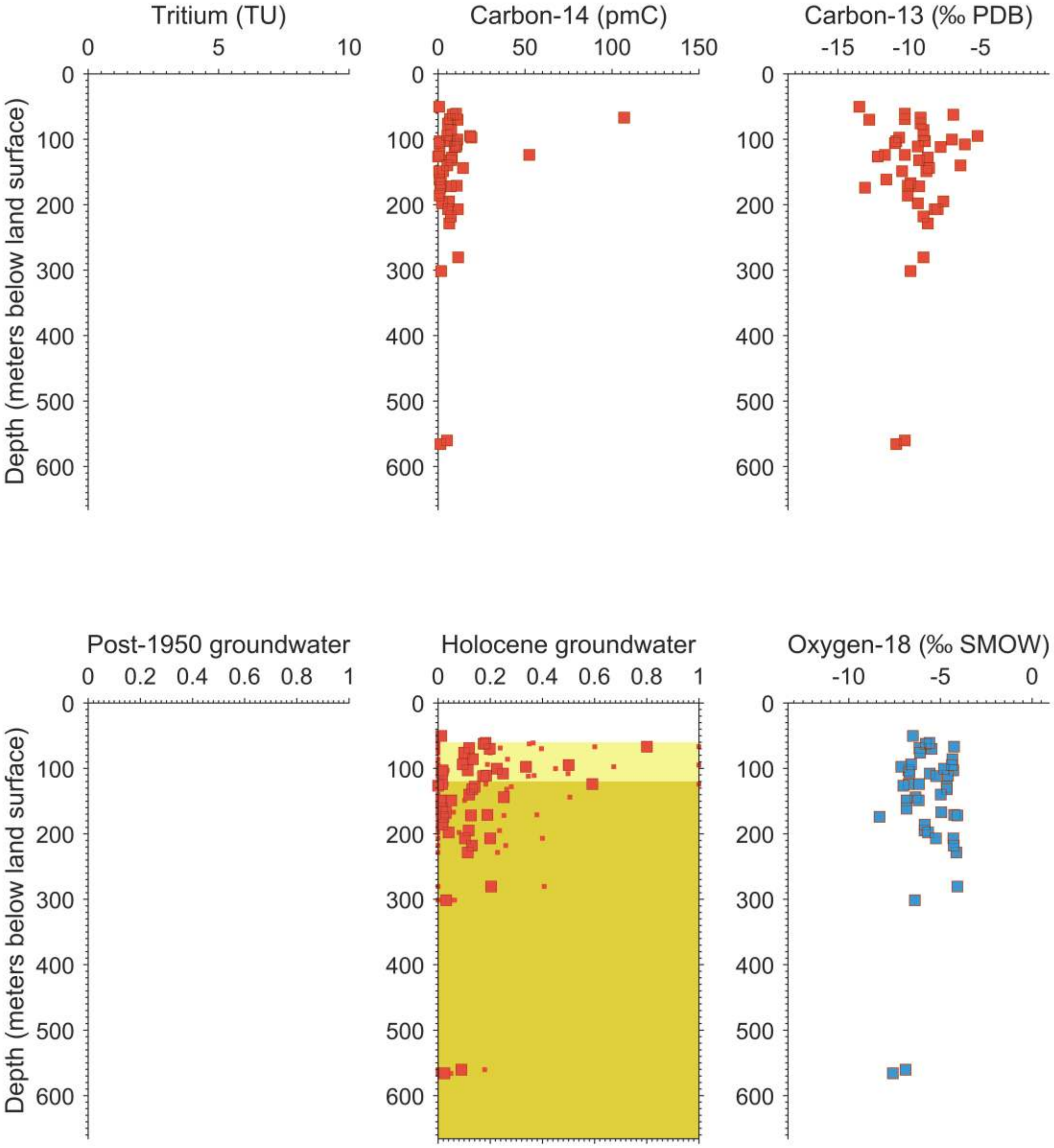
Los Angeles Basin, USA



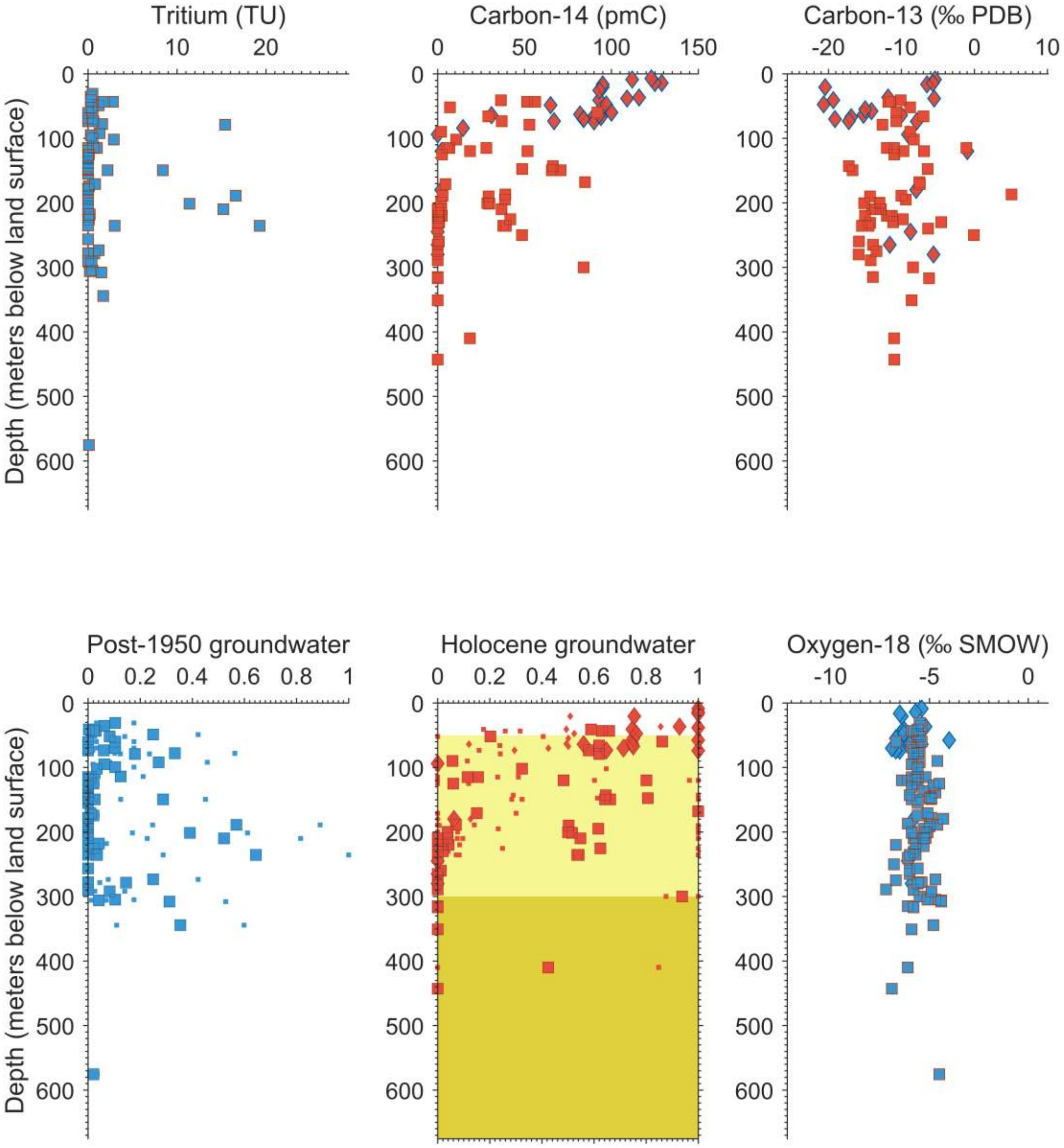
Kazan Trona Ore Field, Turkey (Paleowater transition based on oxygen-18)



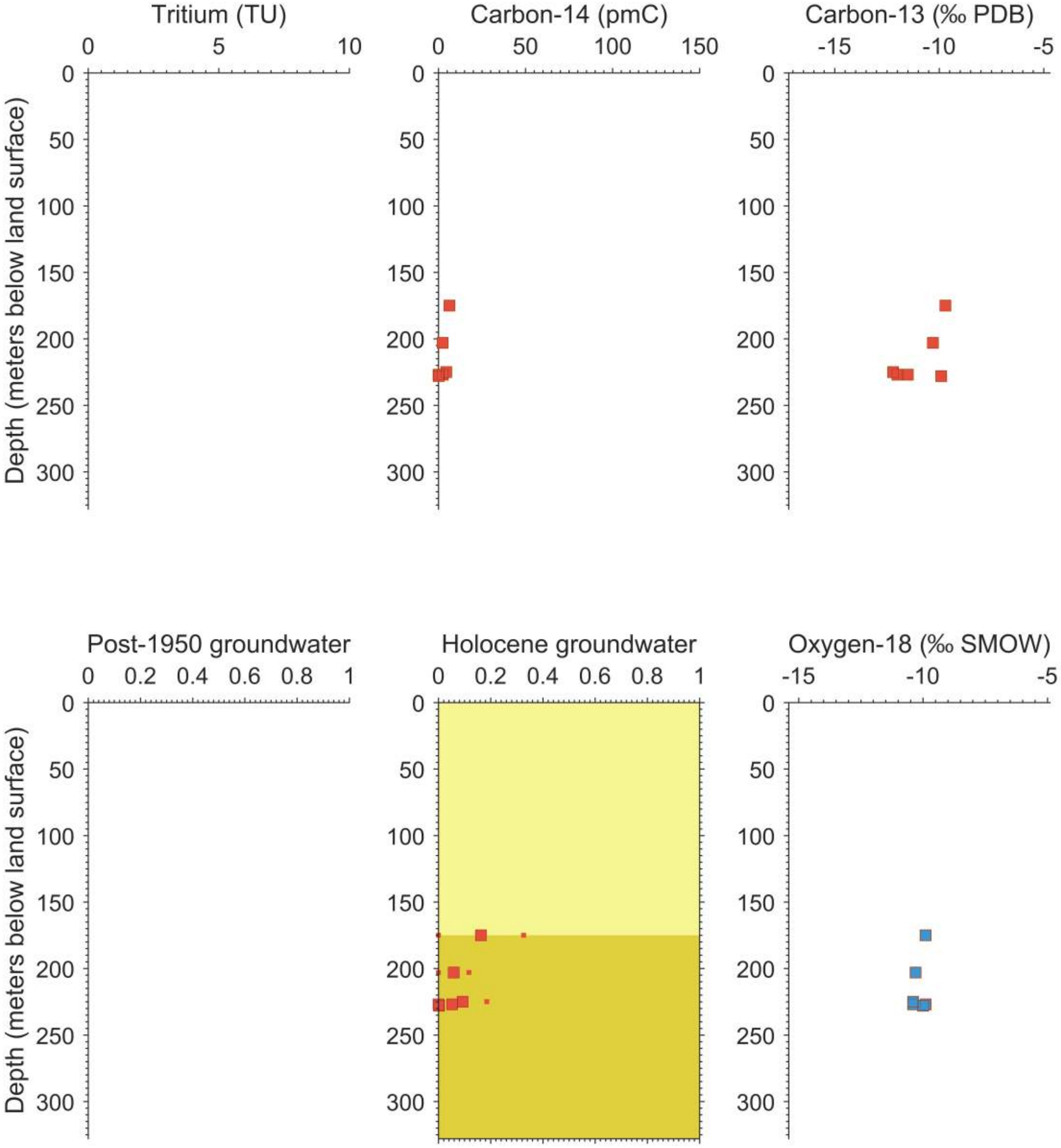
Le bassin du Djérid, Tunisia (Paleowater transition based on oxygen-1



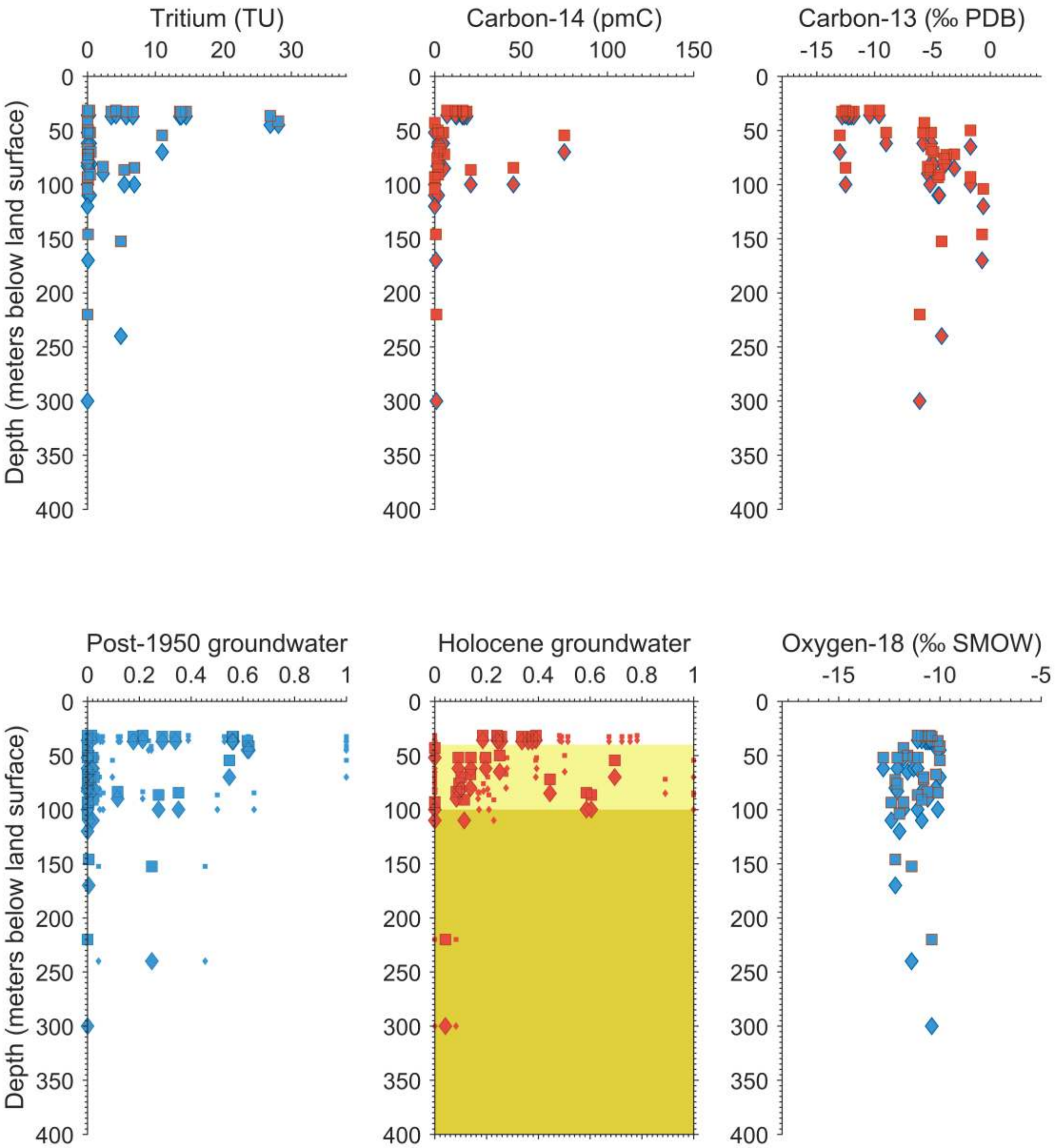
Bangkok Basin, Thailand



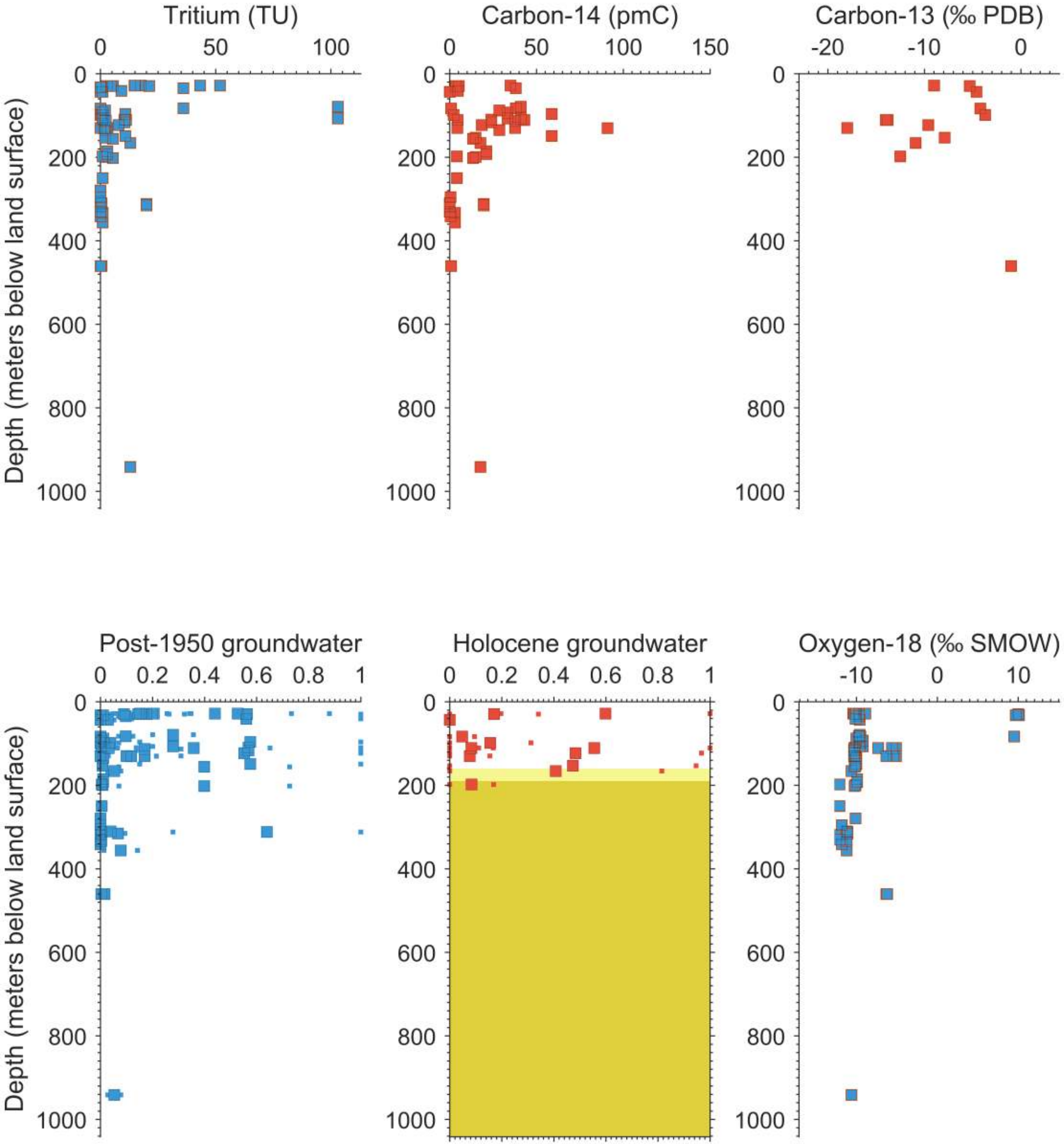
Diass aquifer system, Senegal



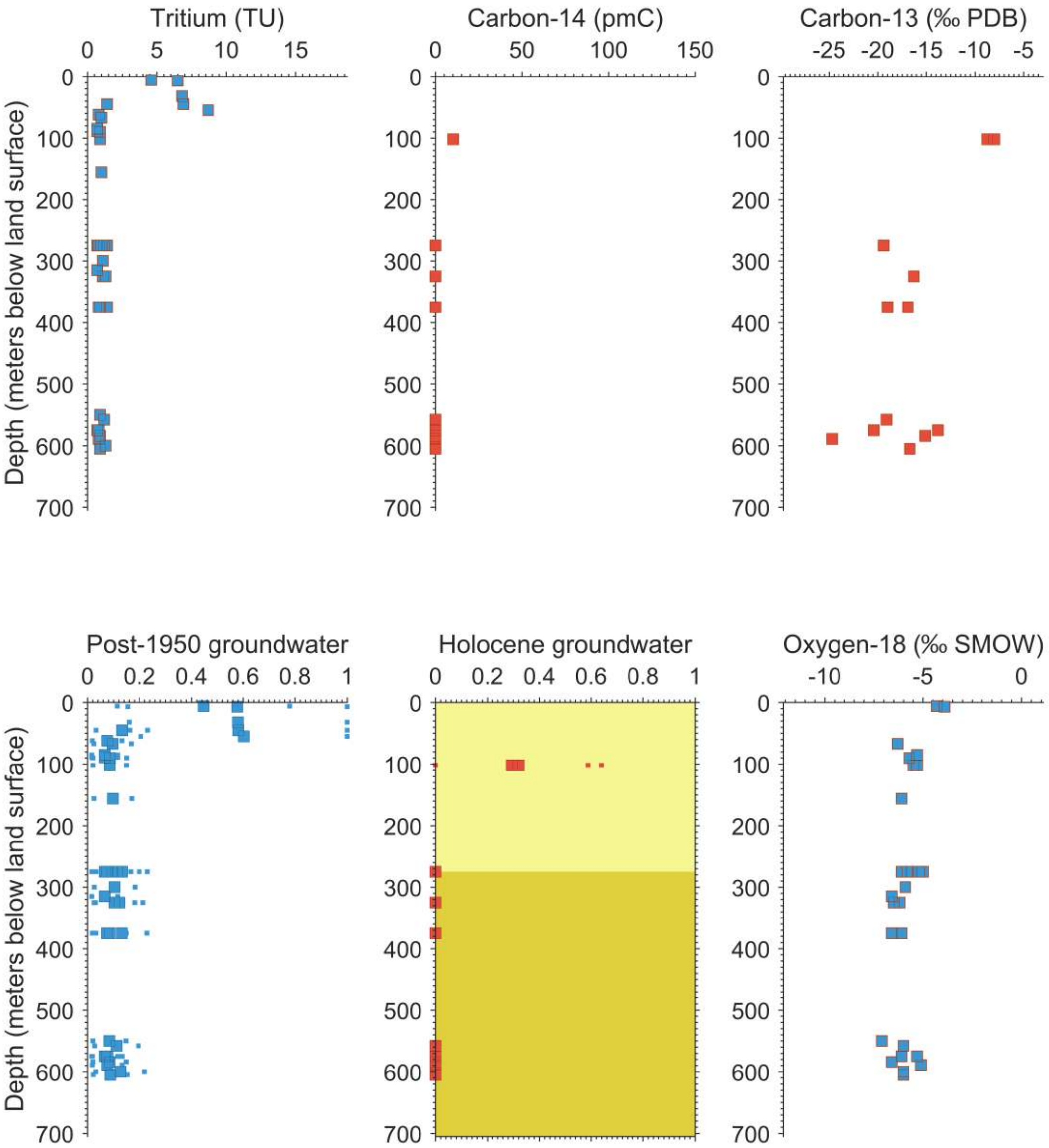
Mazovian basin: Oligocene aquifer, Poland



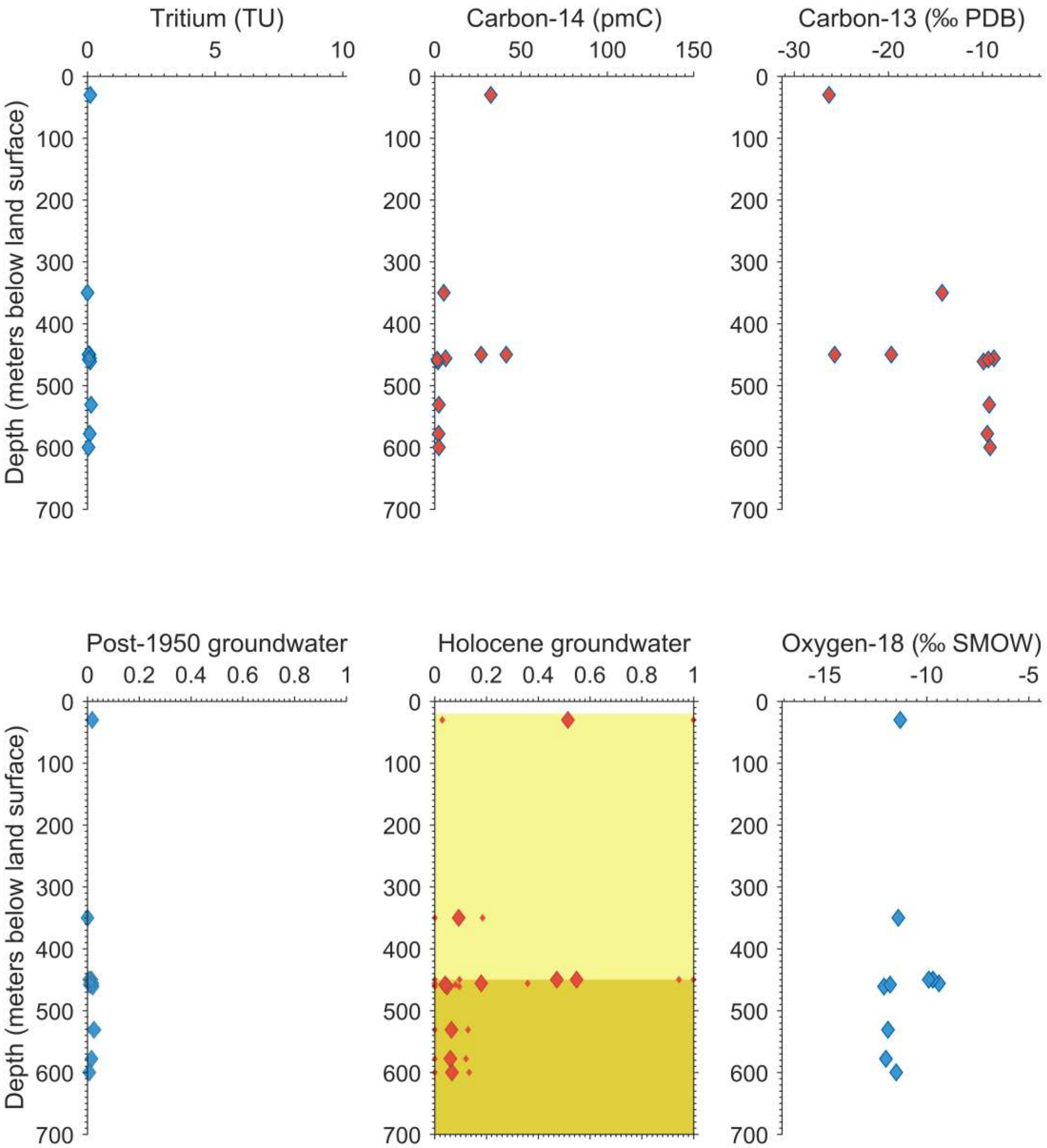
Malm Limestones, Poland



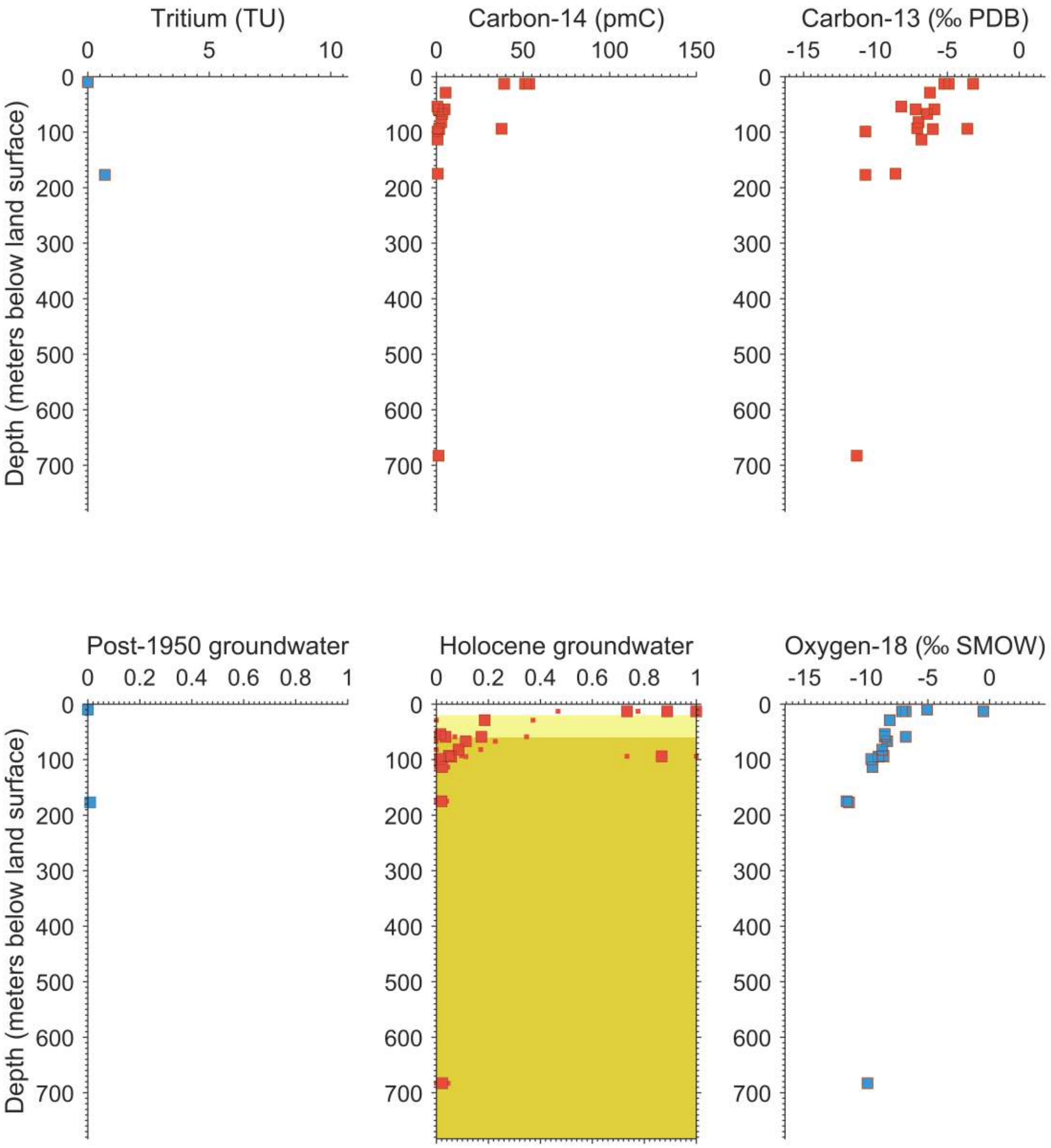
Southern Poland Carbonate Aquifers, Poland



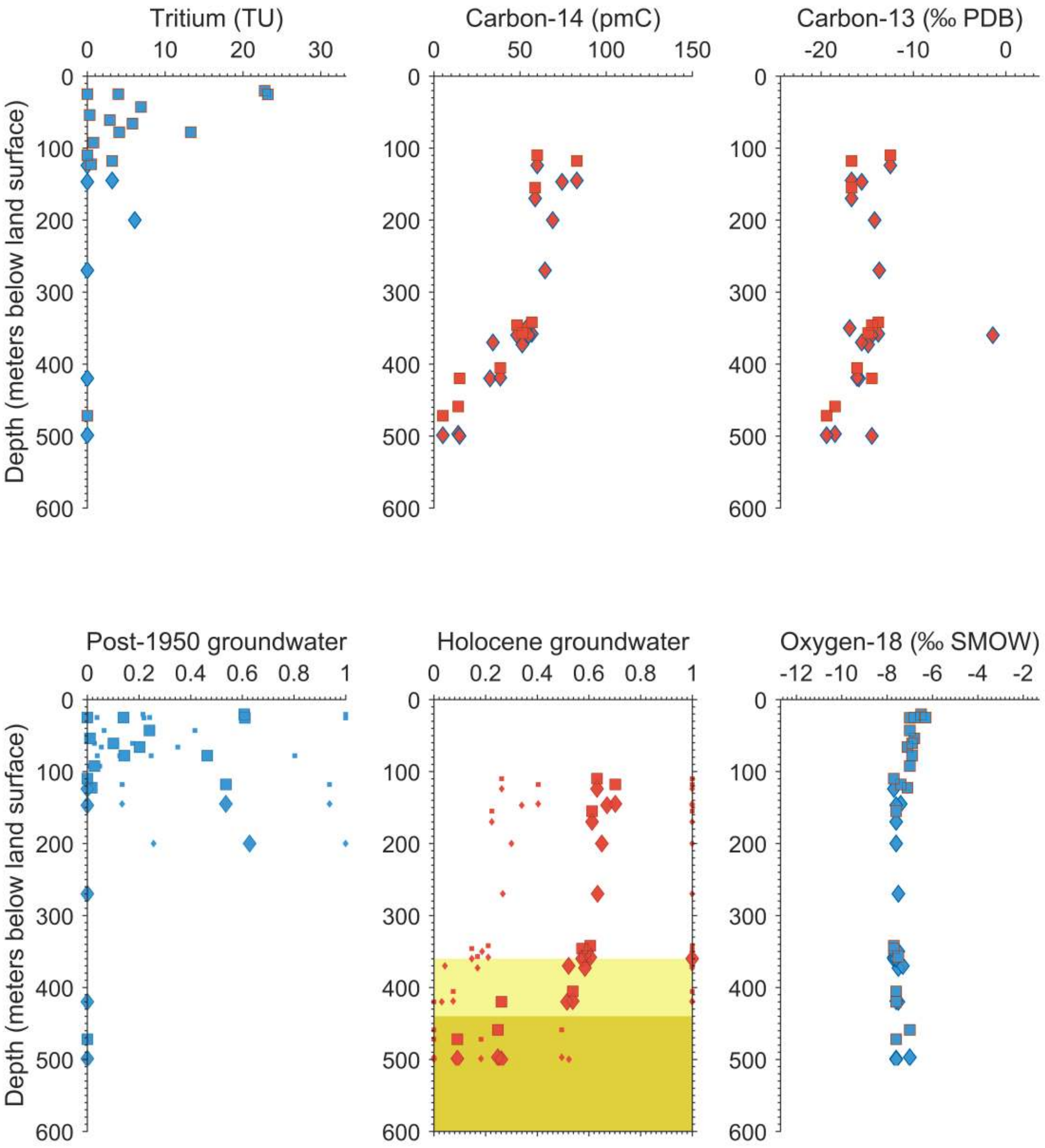
Chad Sedimentary basin (western), Nigeria



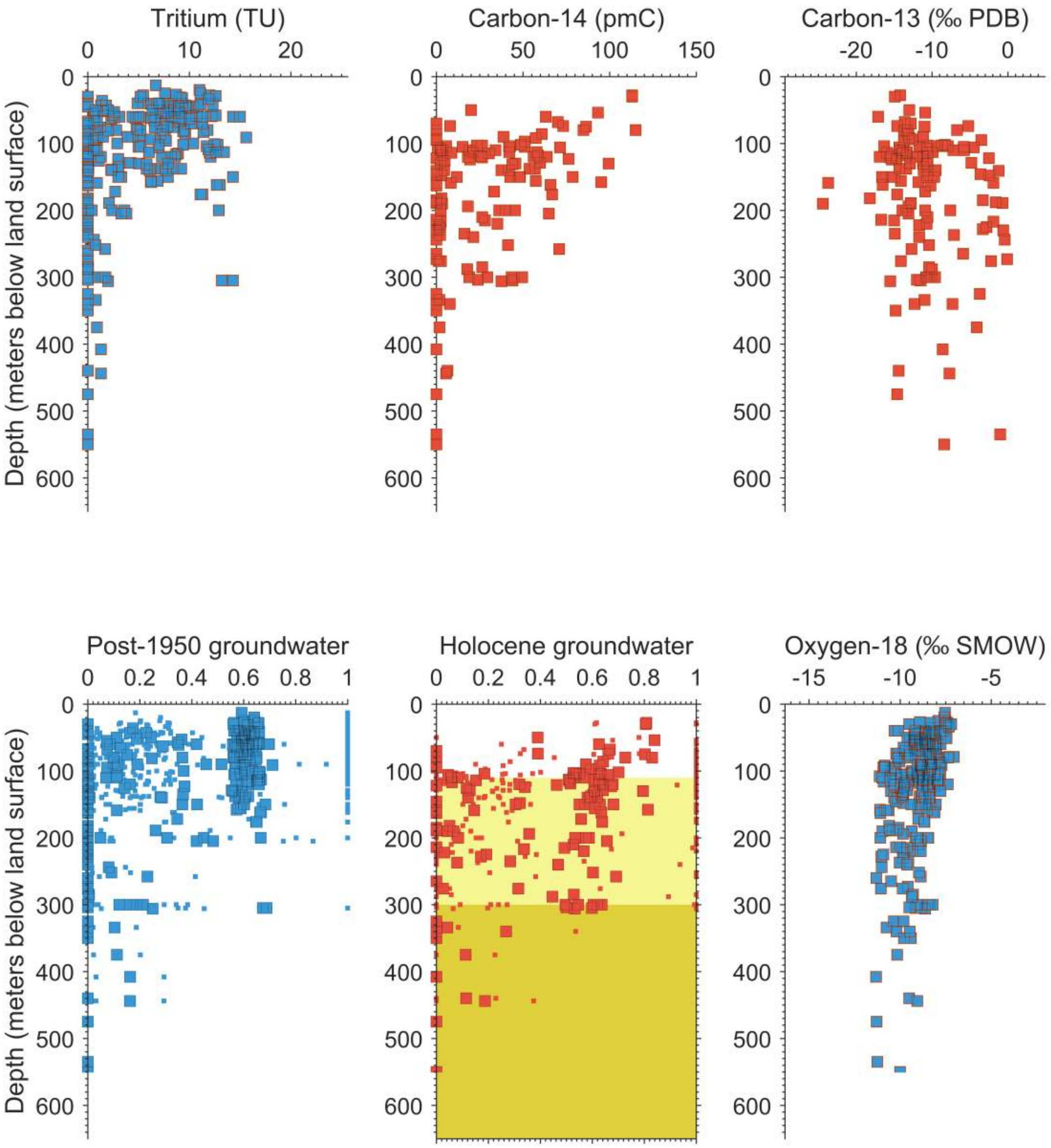
Kufra Basin, Libya



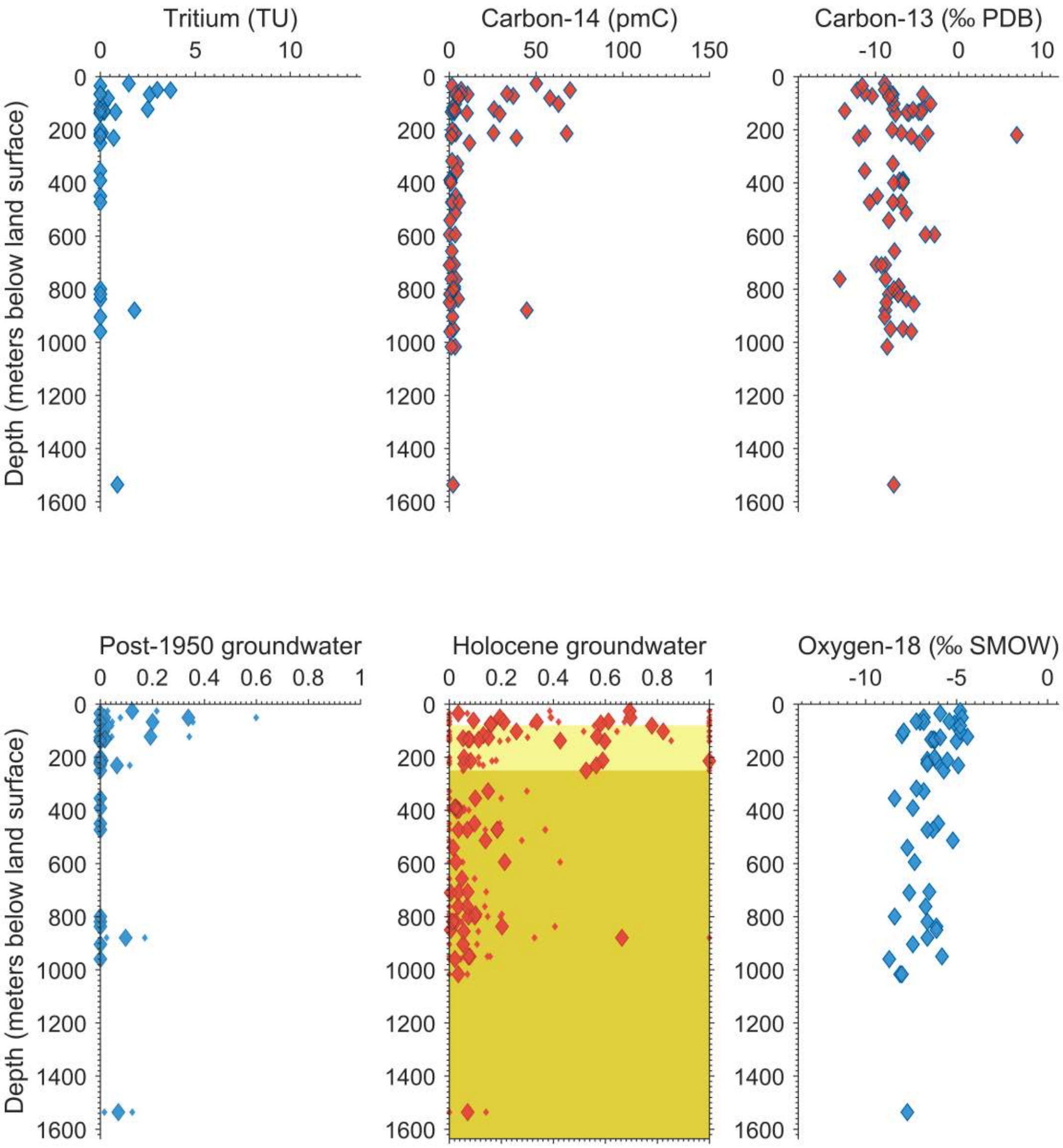
Sirte basin: N Libya, Libya



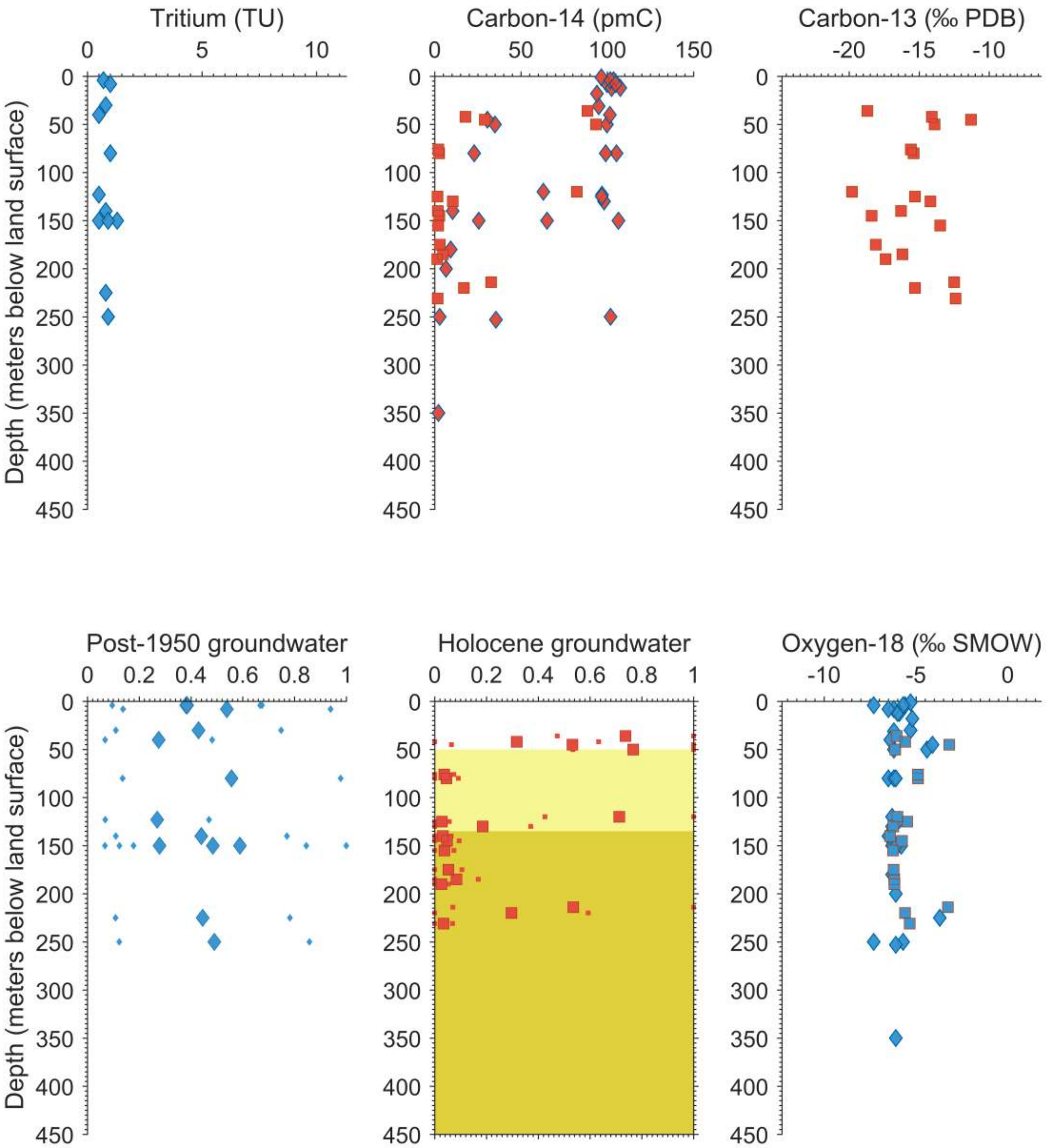
Kimitsu aquifer and Yoro River Basin, Japan



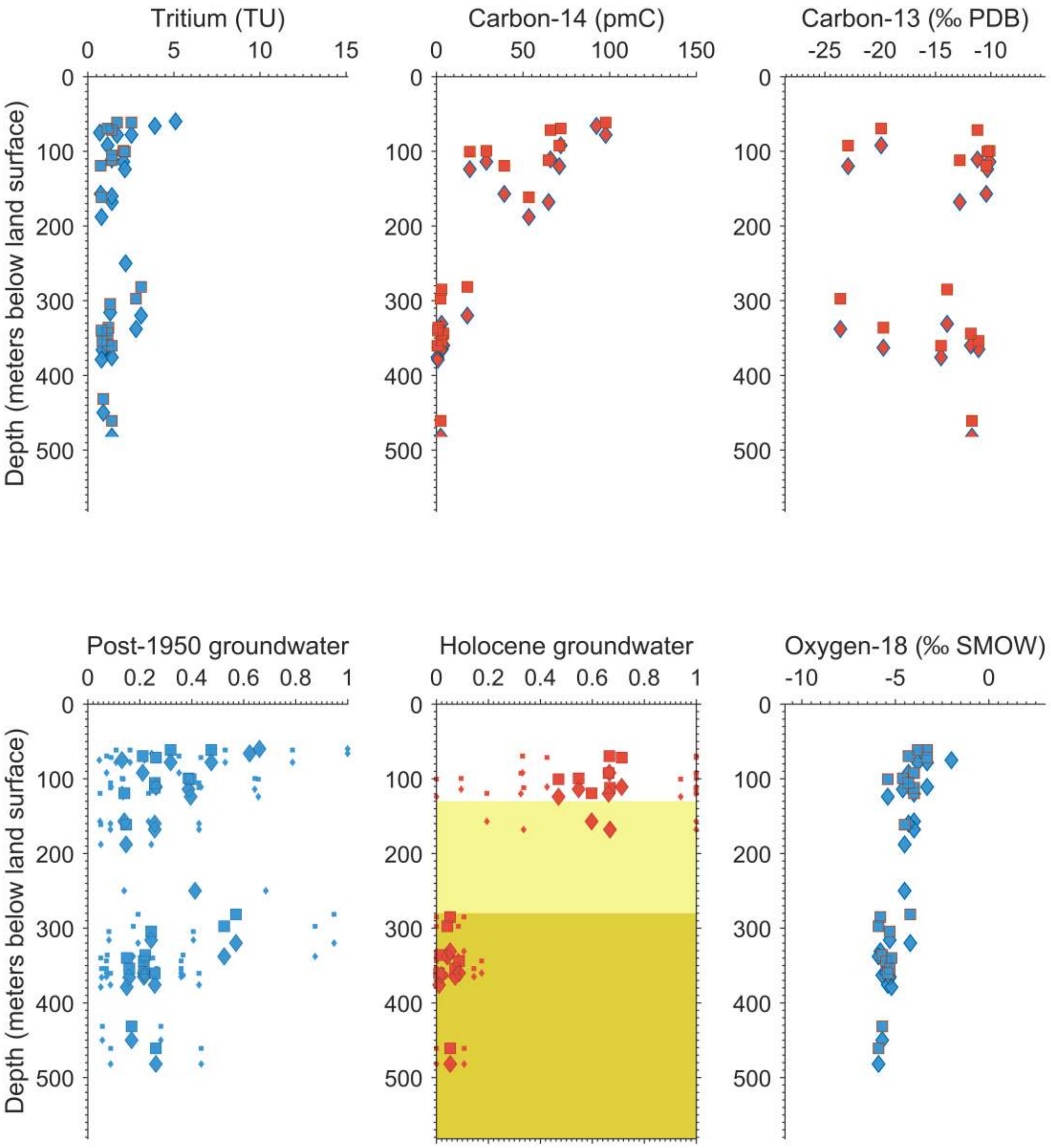
Emilia Romagna plain, Italy



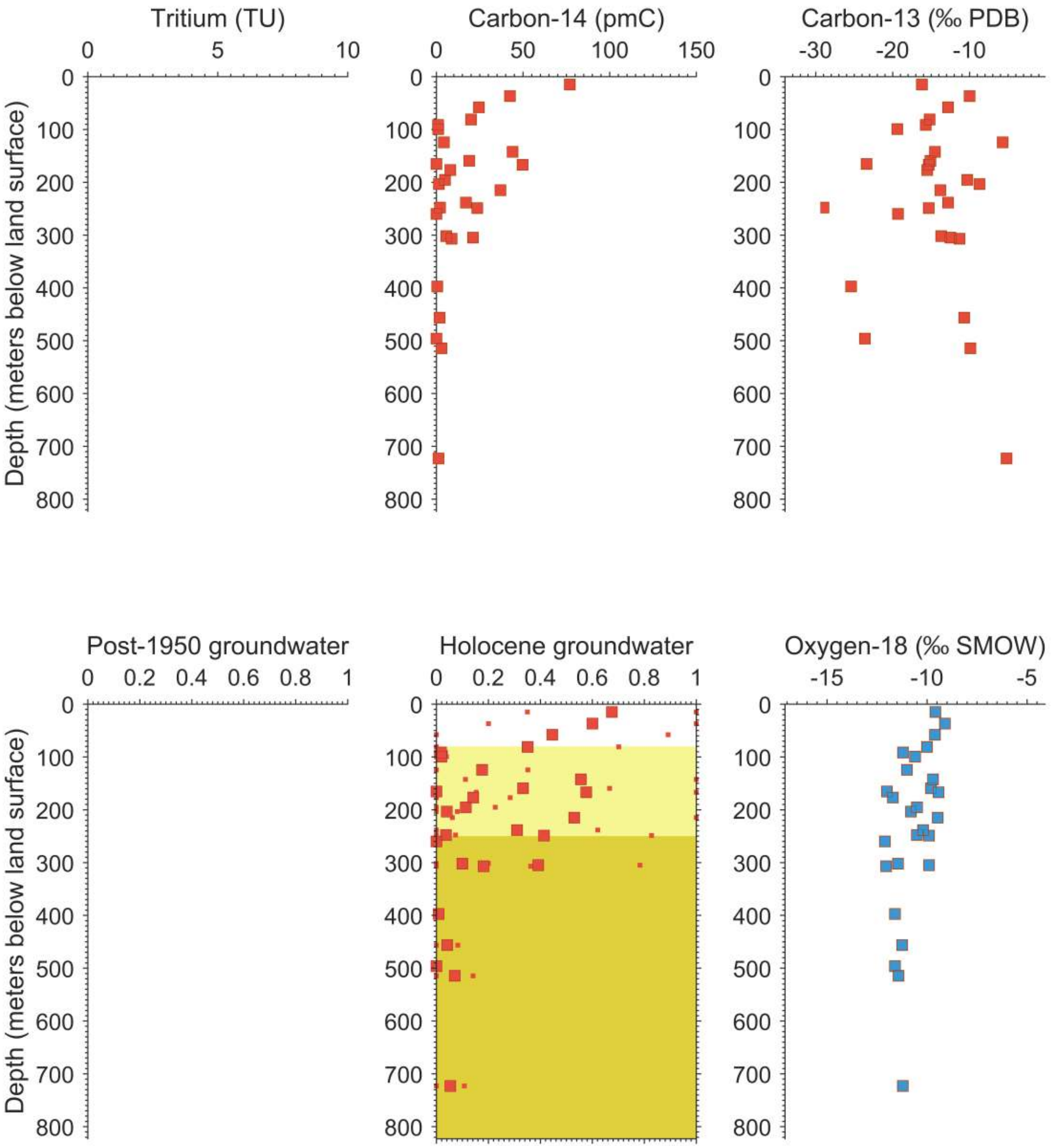
Arava Valley, Israel



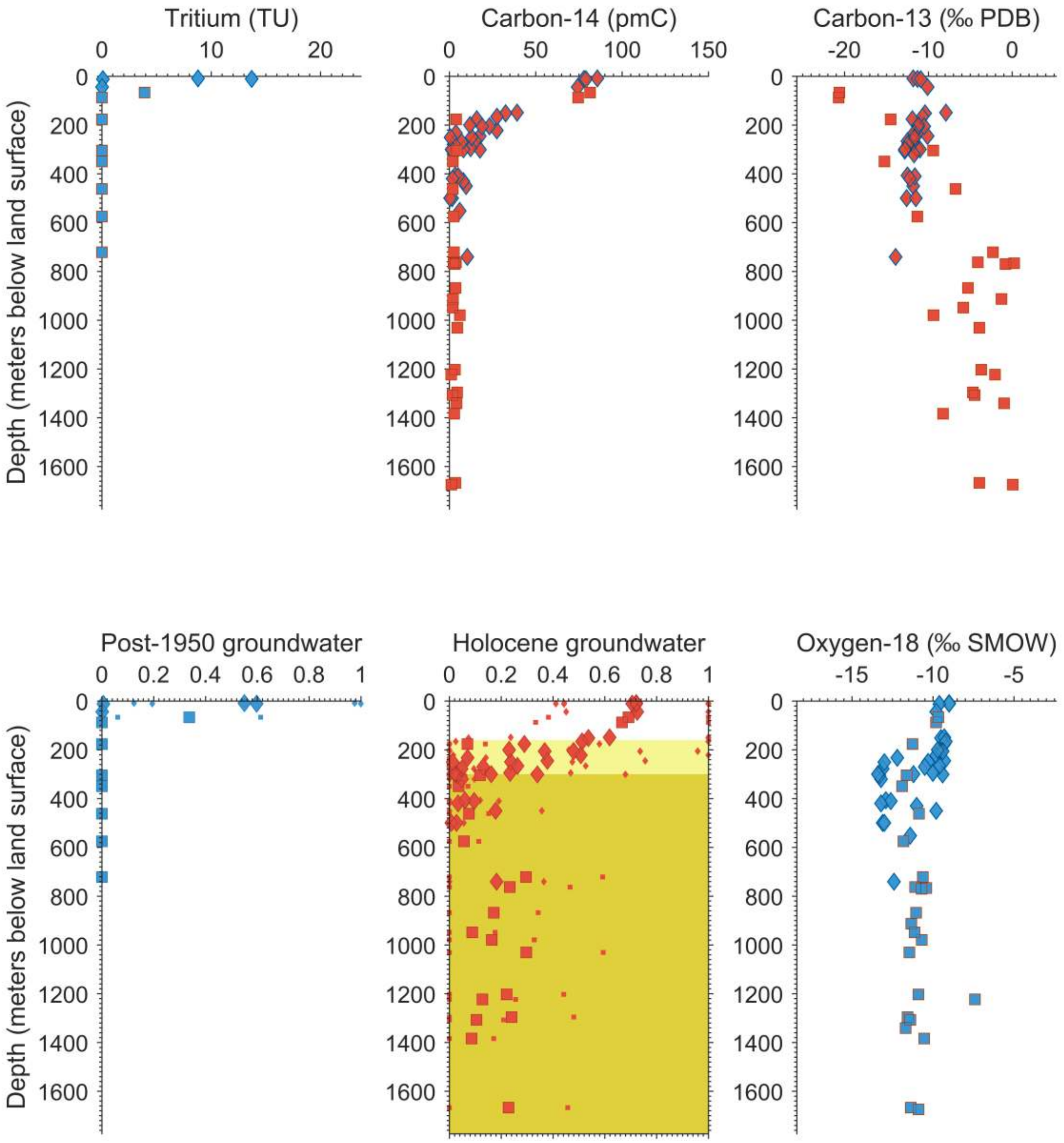
Jakarta basin aquifer, Indonesia



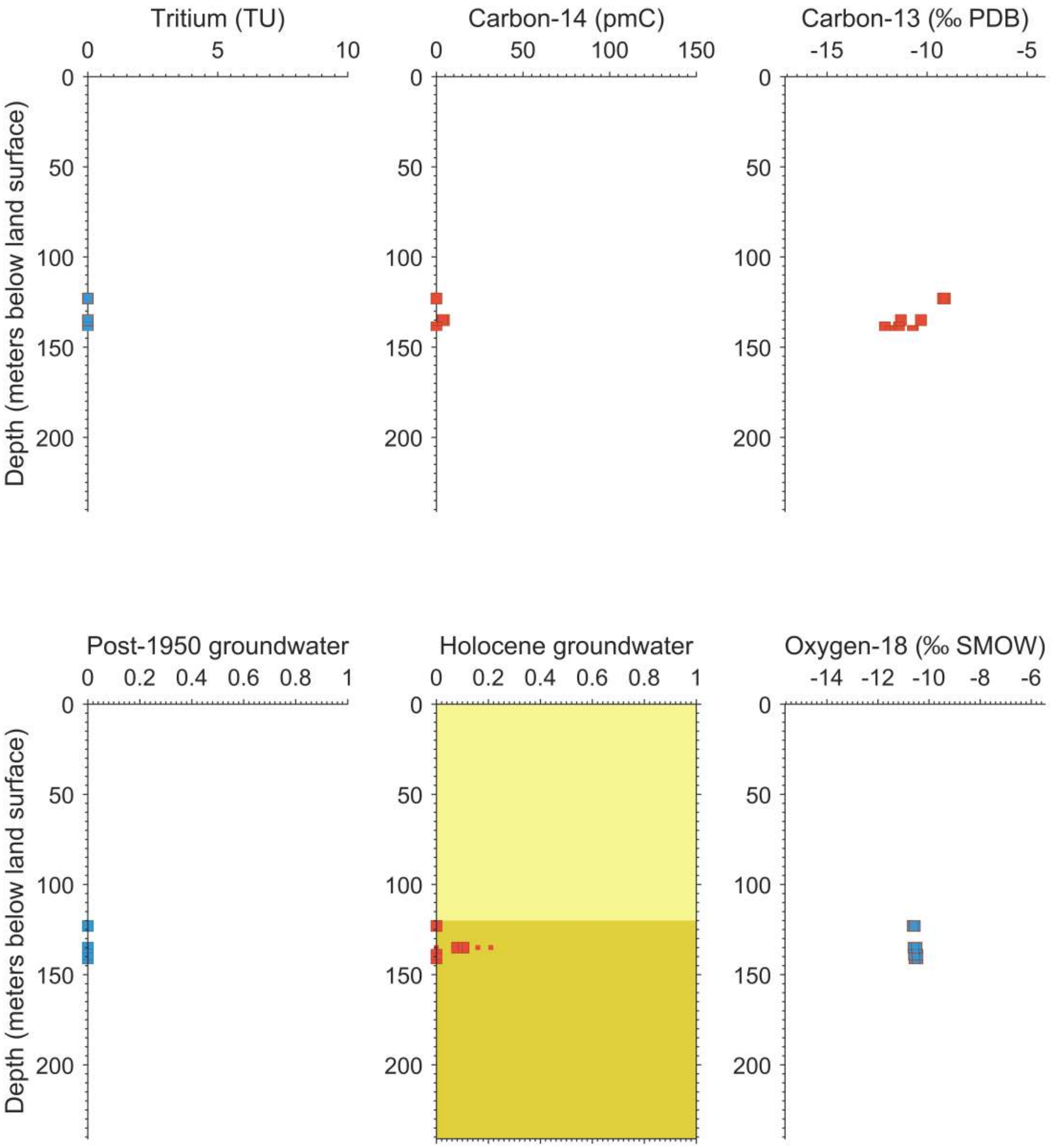
Tiruvadanai aquifer, India



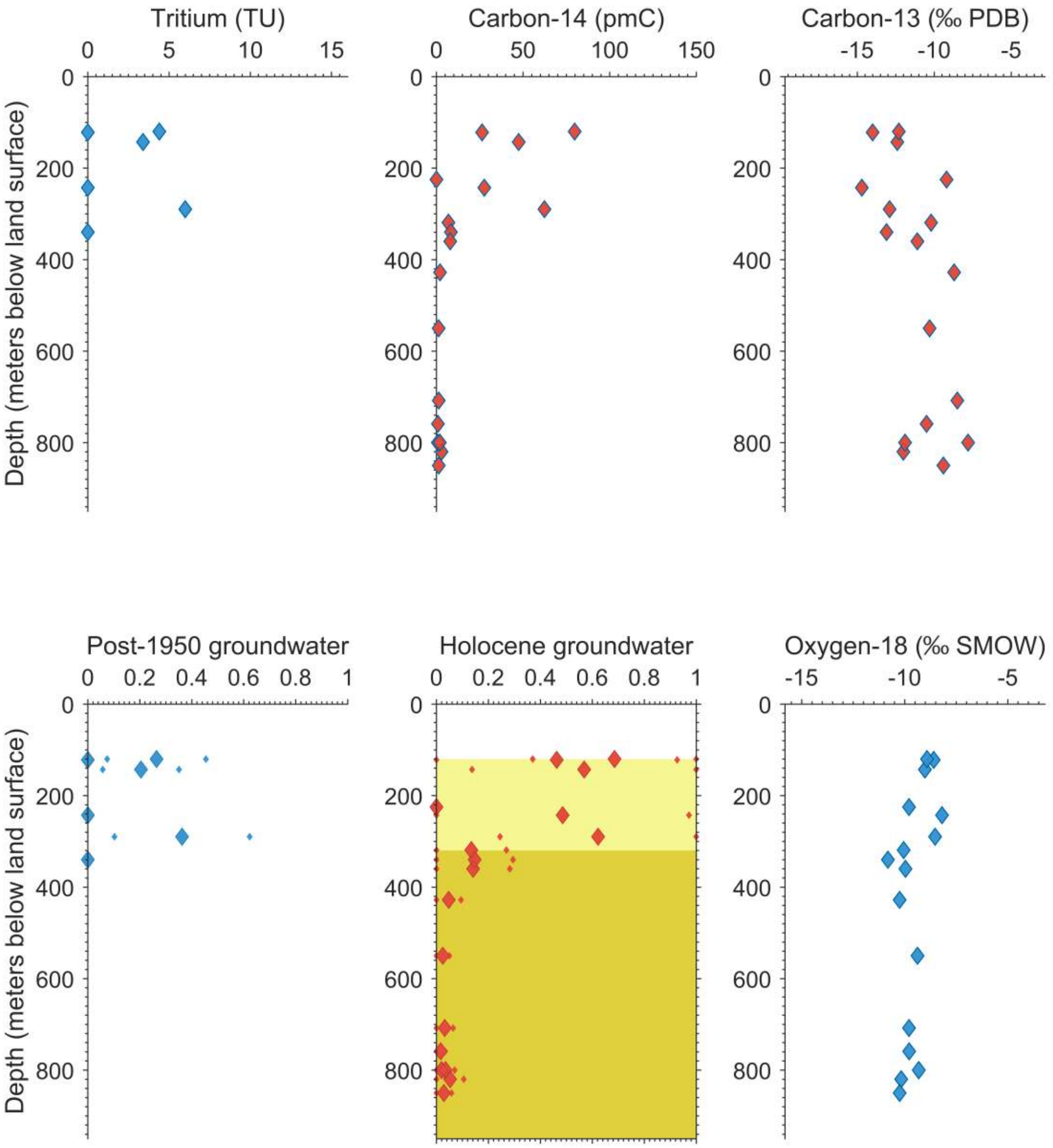
Great Hungarian Plain: Shallow unconfined, Hungary



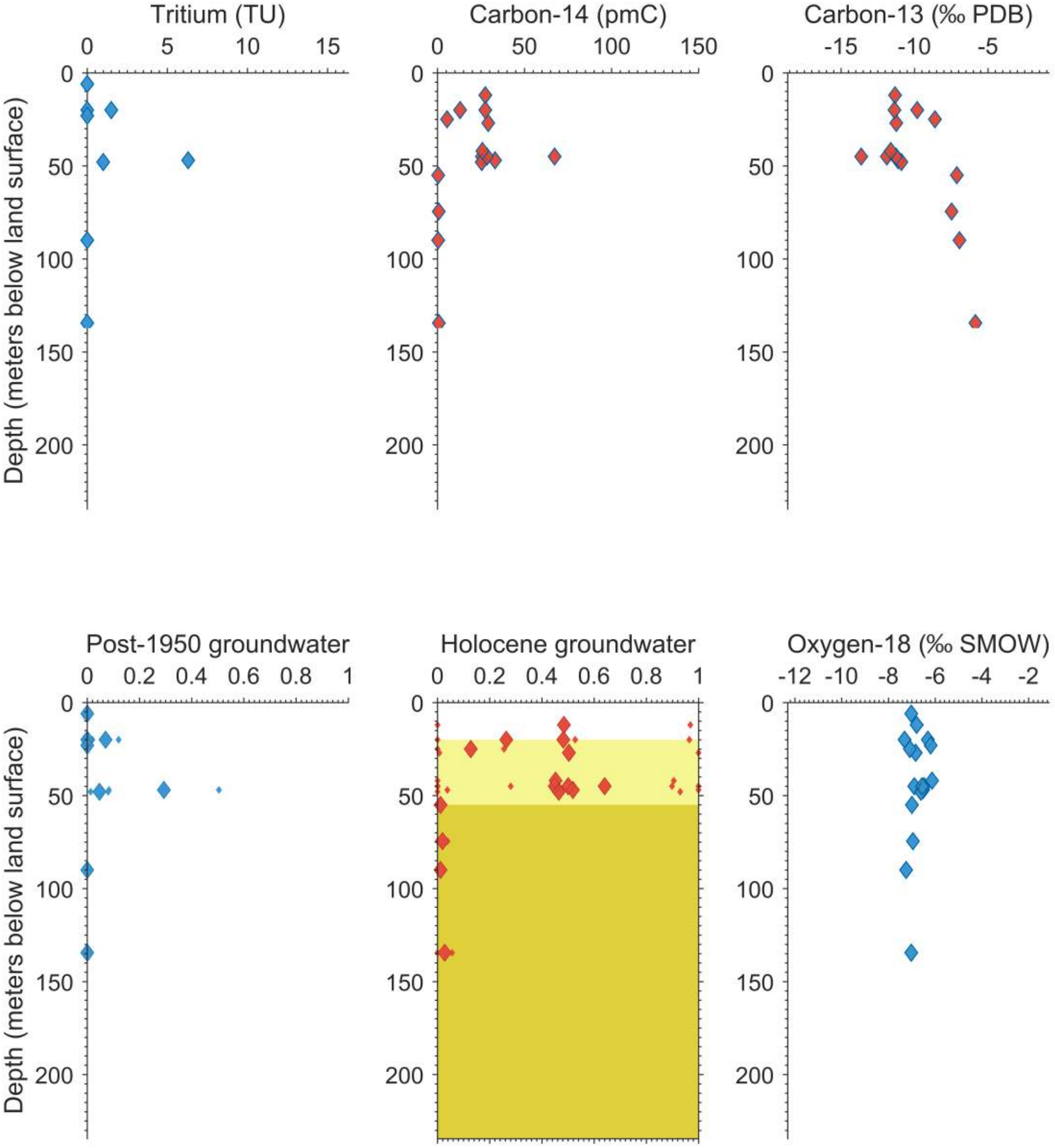
Pannonian Basin, Hungary



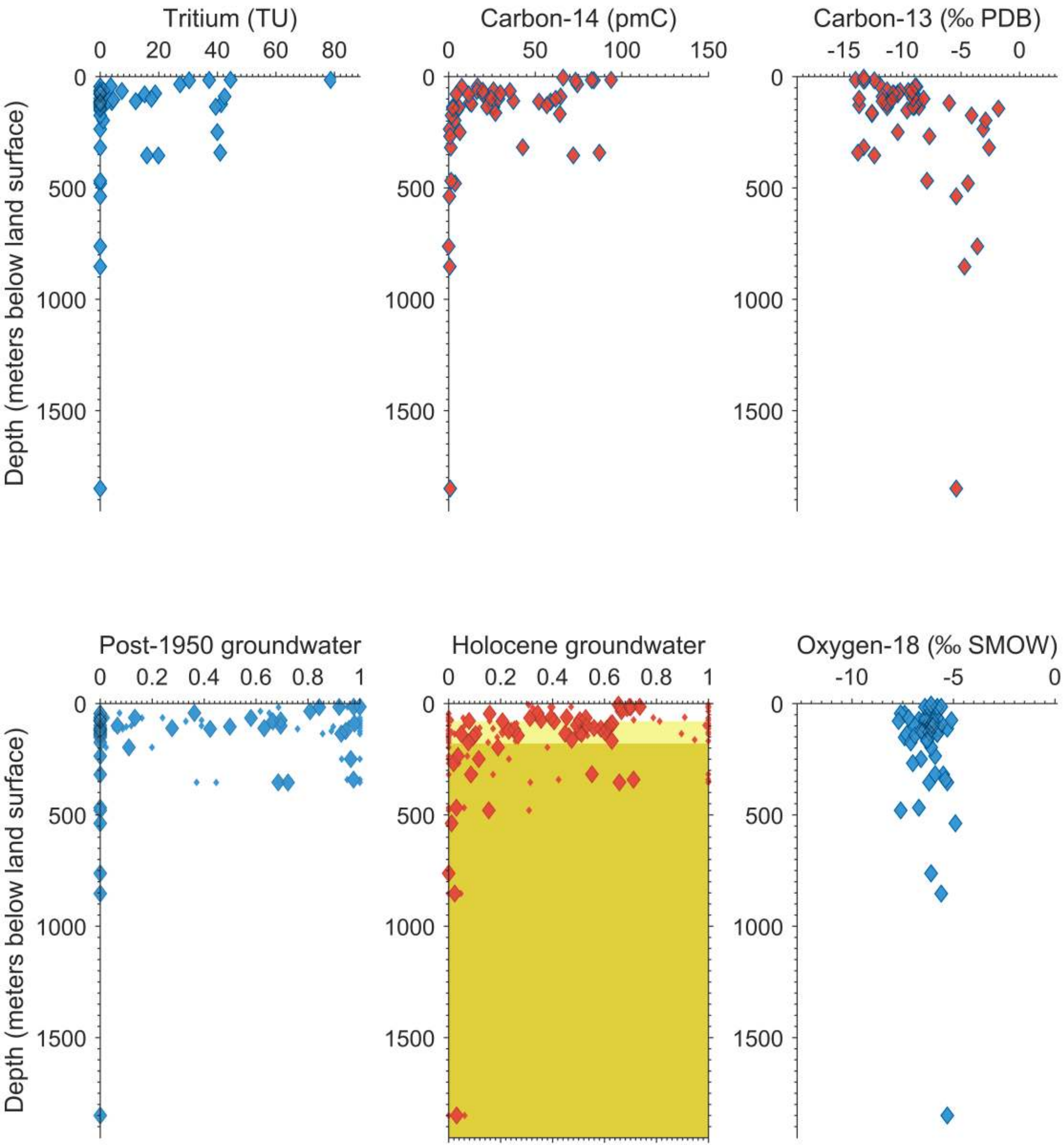
Benkerstein, Germany



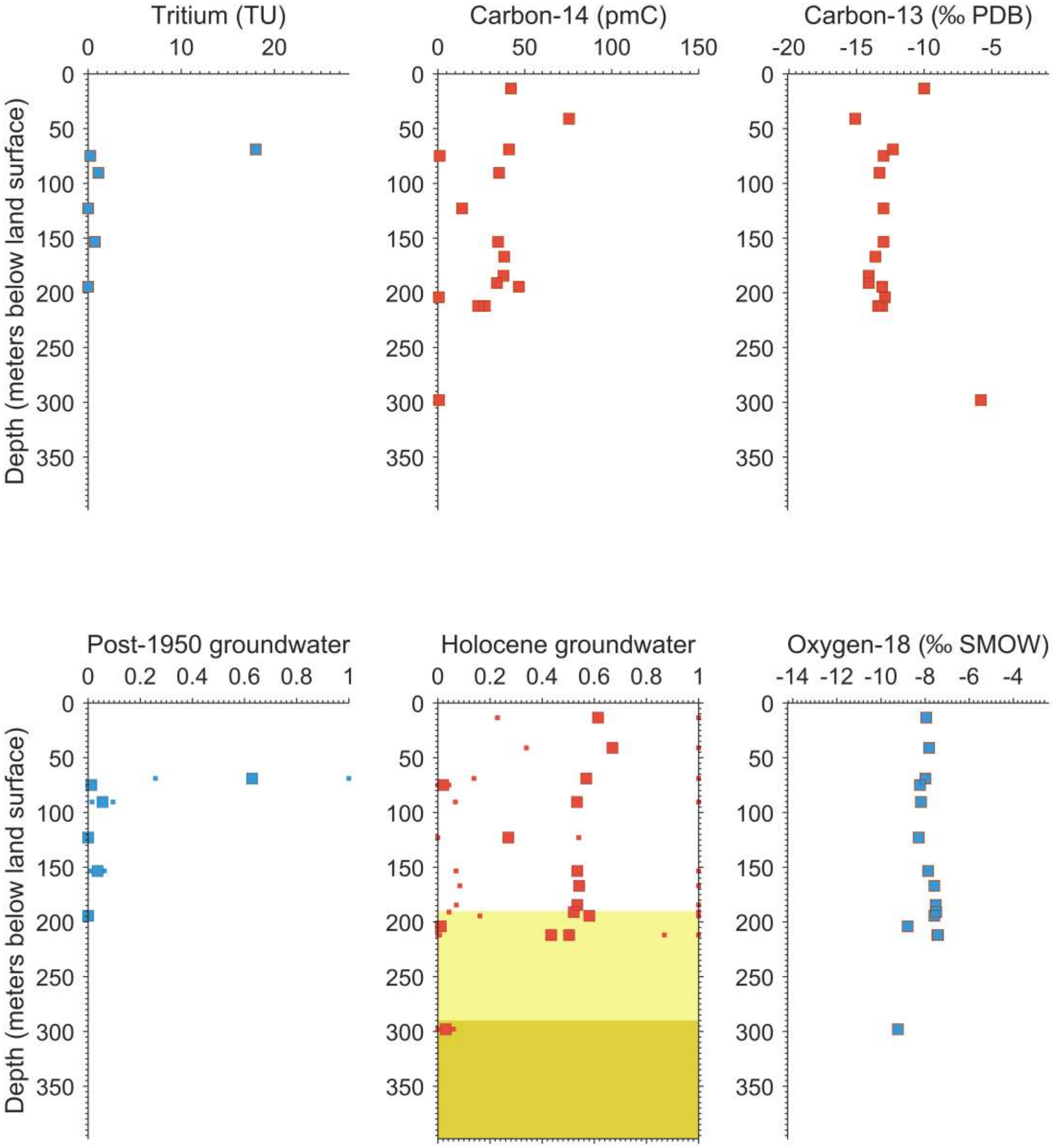
Lorraine Sandstone Aquifer, France



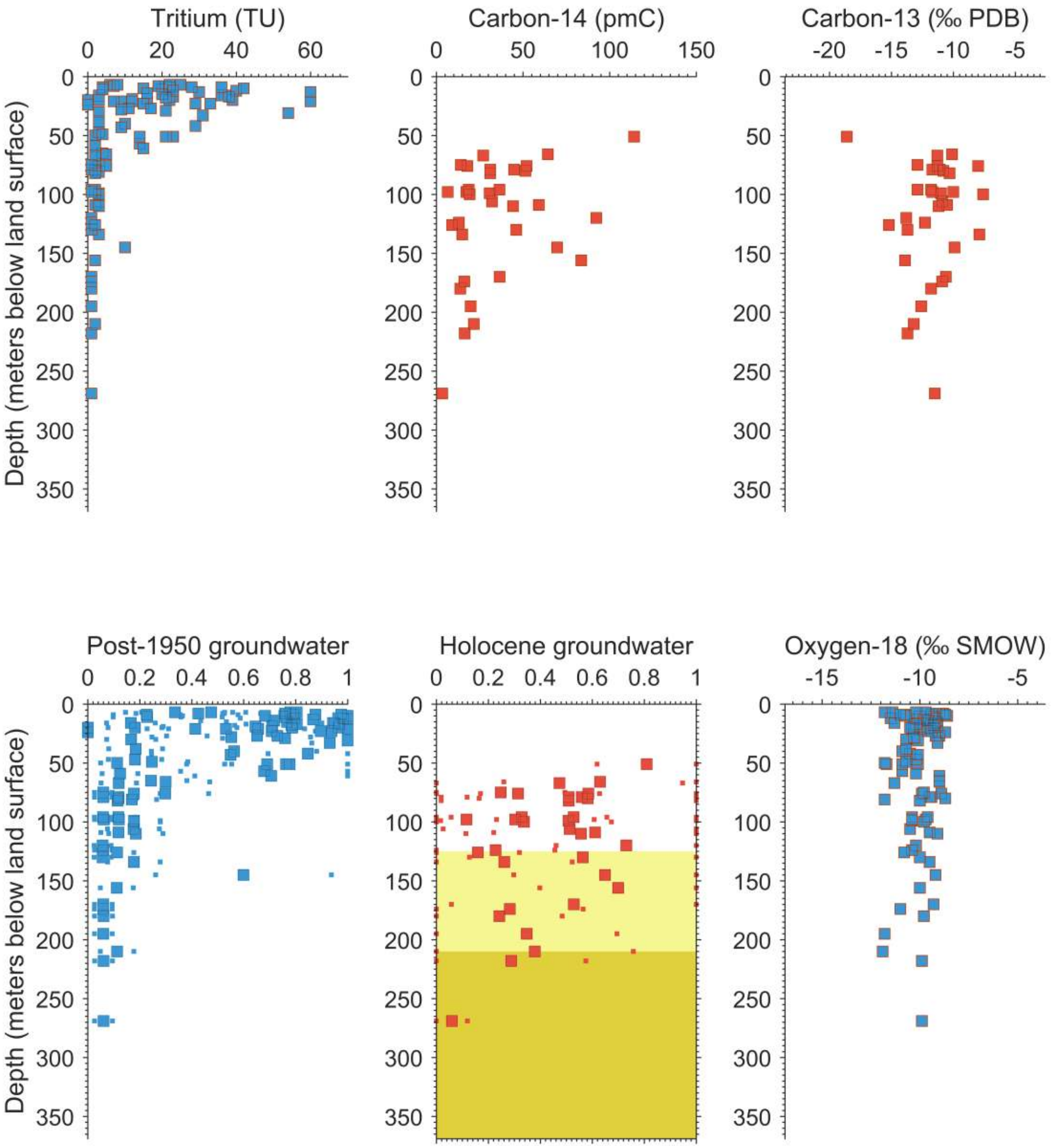
Bathonian and Bajocian coastal aquifer, France



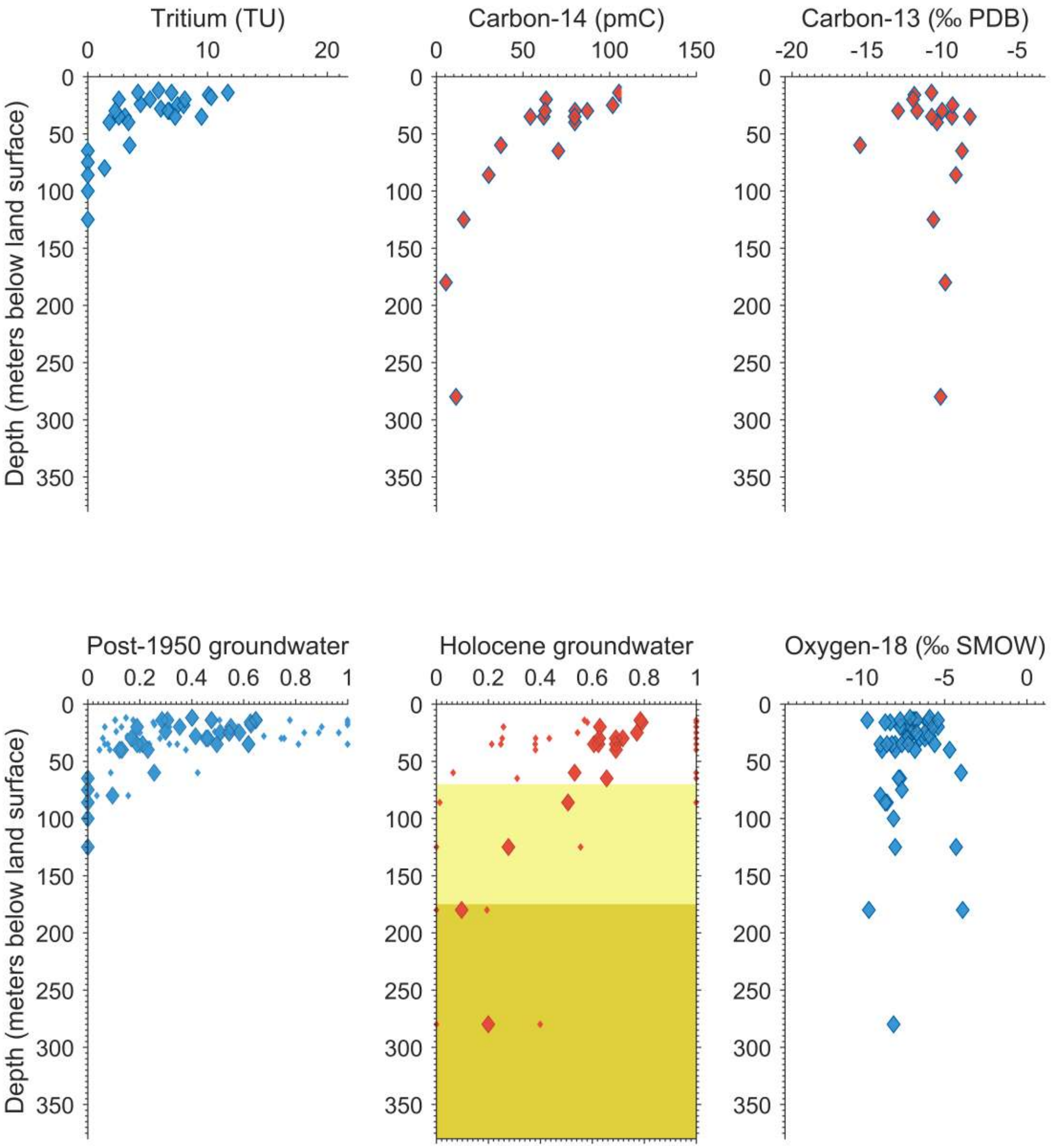
Aquitaine Basin, France



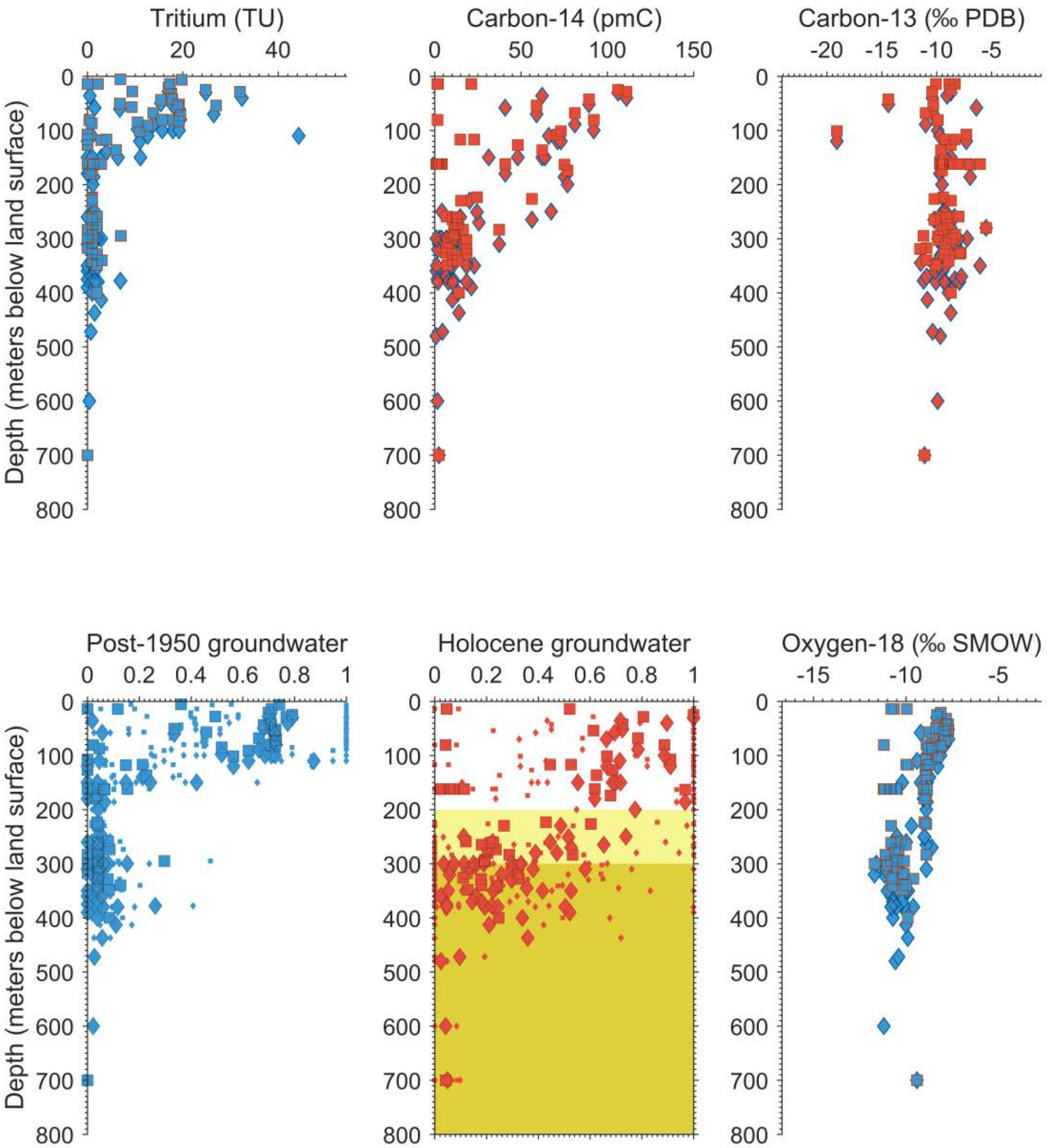
Ribe Formation, Denmark



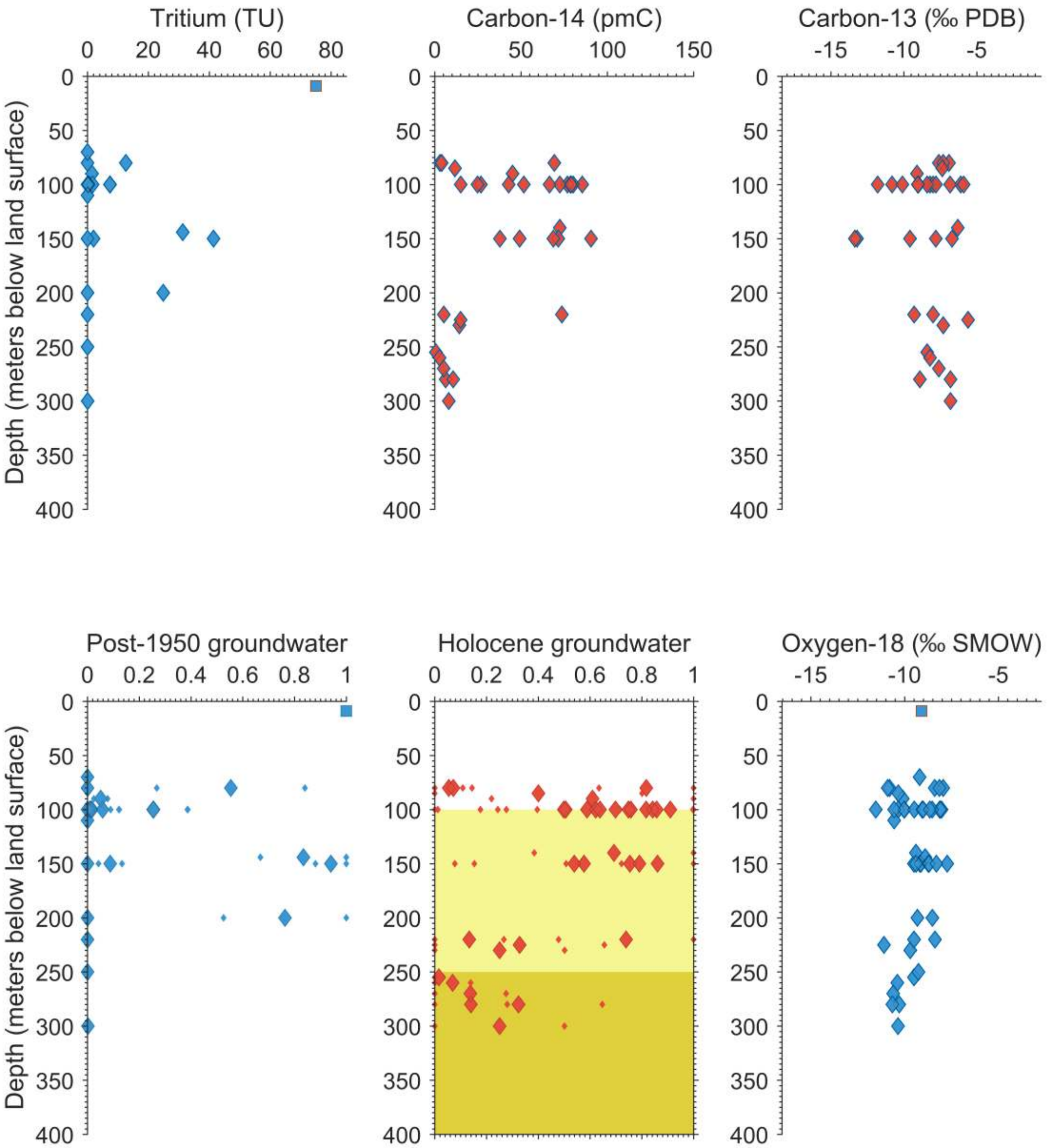
Songnen Plain aquifer, China



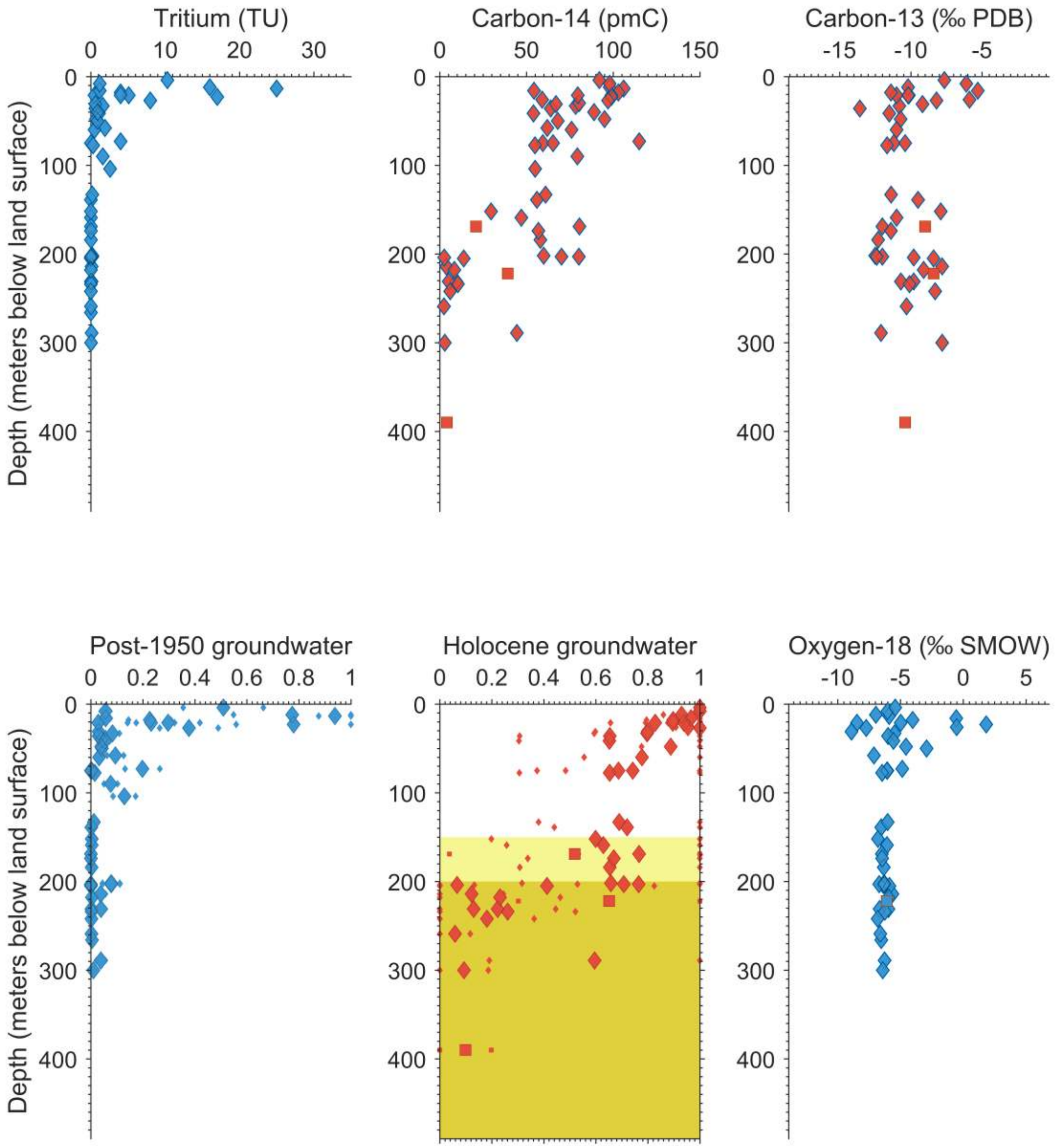
Laizhou Bay, China



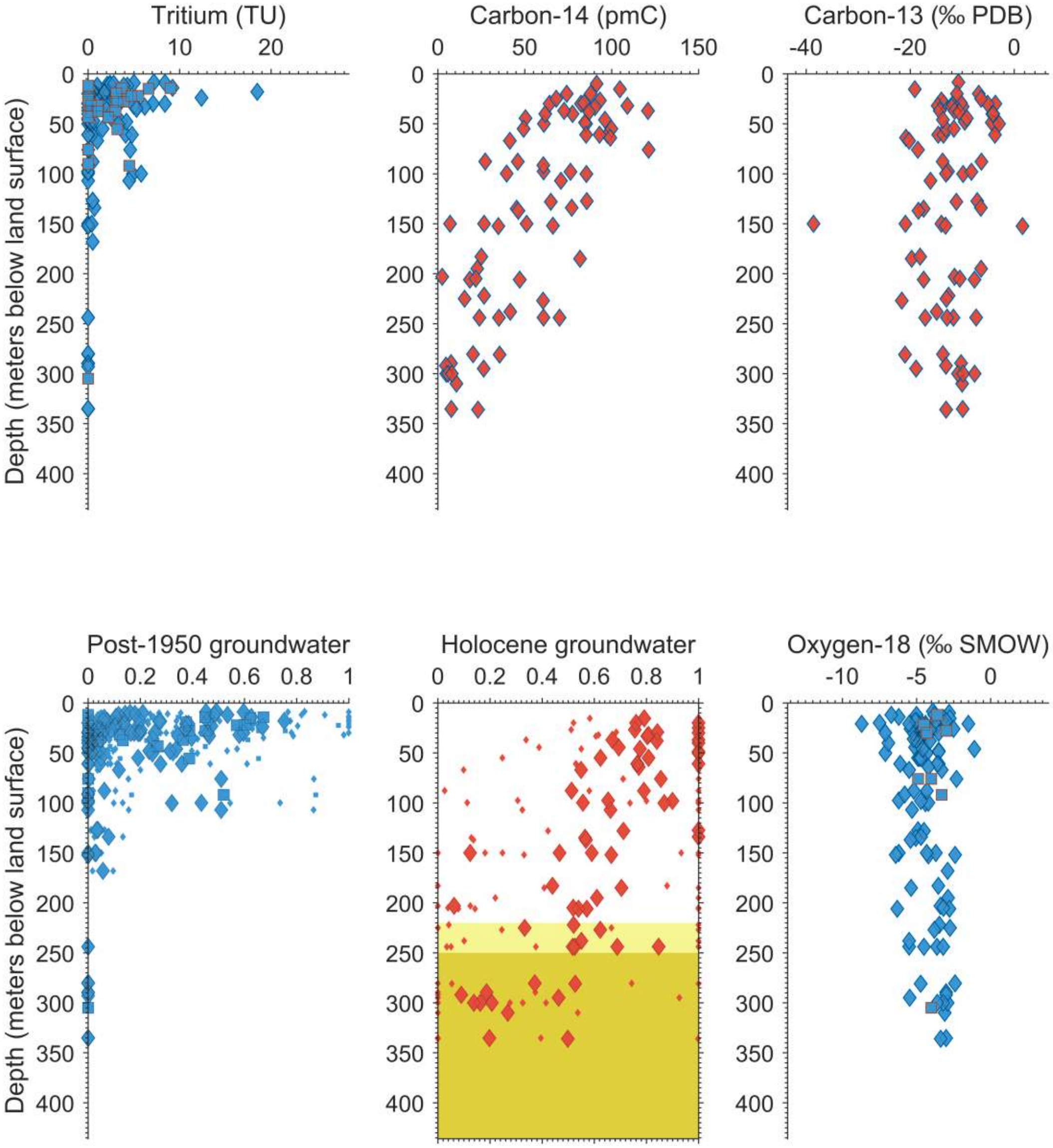
North China Plain, China



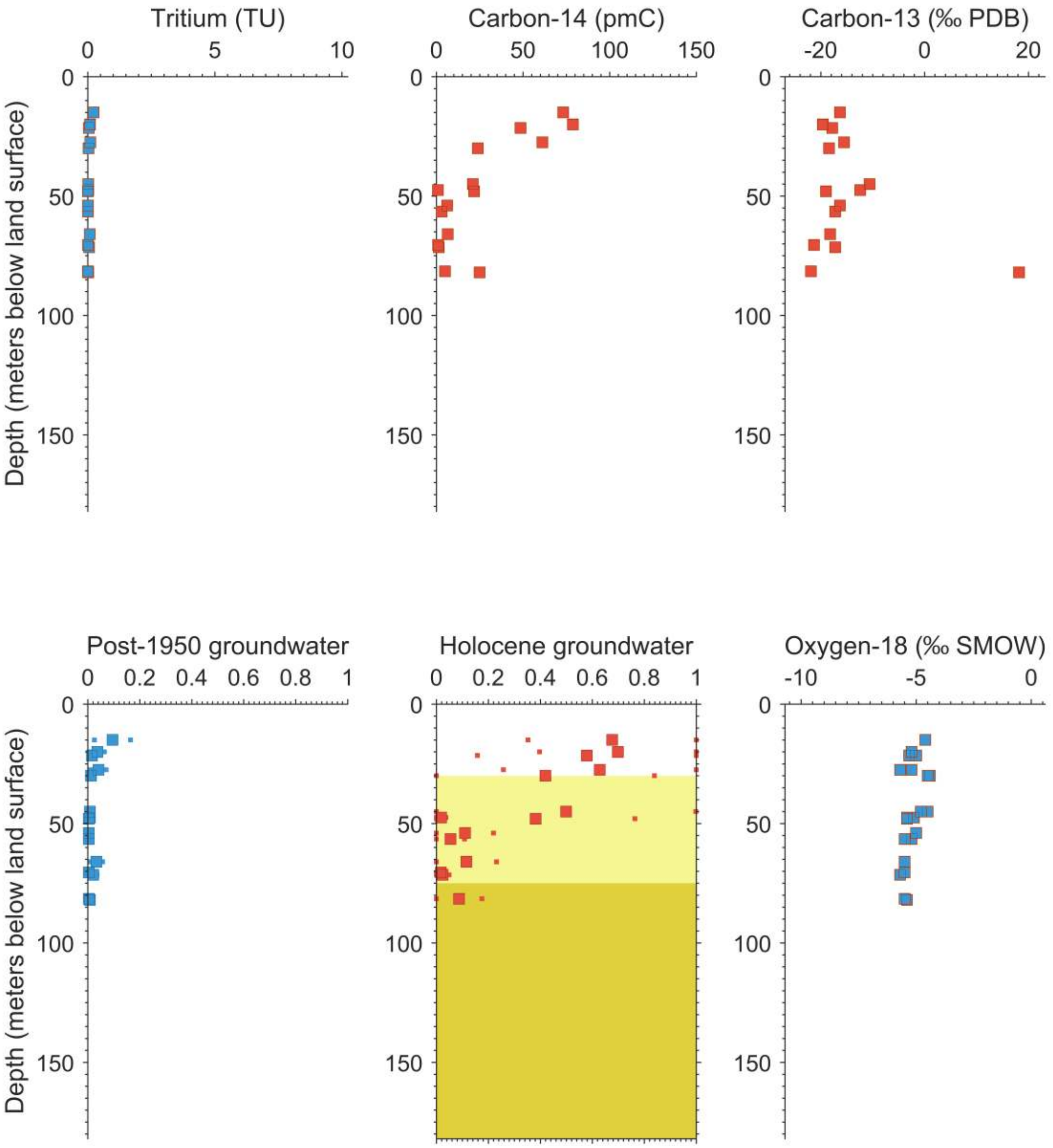
Eastern Hexi Corridor, China



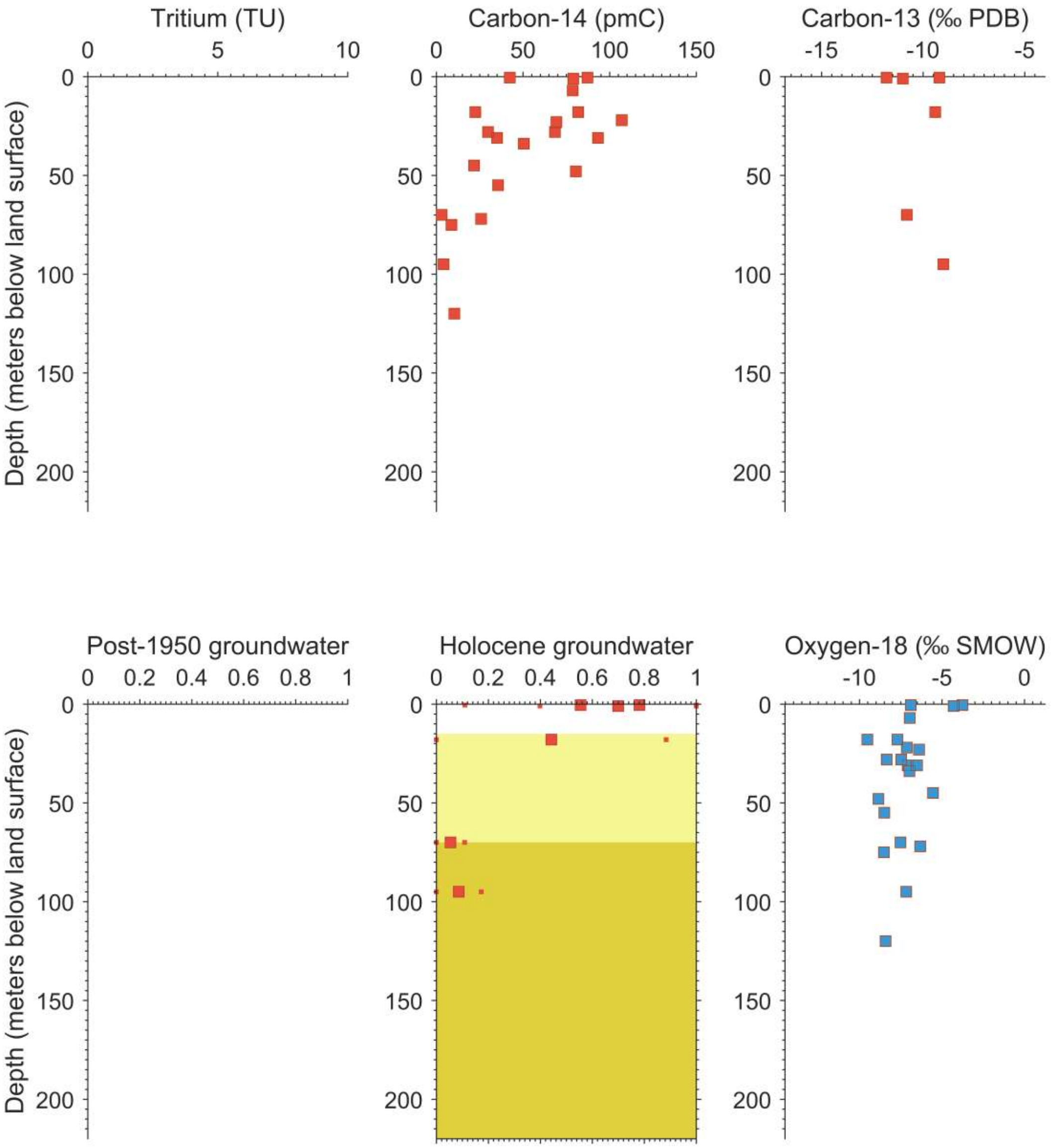
Kalahari Desert: Ntane Sandstone, Botswana



Bengal Basin, Bangladesh



Western Port Basin, Australia



Ngalia and Amadeus Basin, Australia