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MINI-REVIEW

In Danger One of the Largest Aquifers in the World, the Great Mayan Aquifer, Based on Monitoring the Cenotes of the Yucatan Peninsula

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Abstract
The aquifer flowing beneath the Yucatan Peninsula, México, is one of the largest in the world and is in direct contact with the surface through "cenotes" (sinkholes) that have been documented to be contaminated with various classes of pollutants. The objective of this study was to evaluate the environmental status of the Great Mayan Aquifer through a review of data published on pollution of the cenotes. Approximately 1000 known georeferenced cenotes on the Yucatan Peninsula were geographically located. A map was generated using the geographic information system software. High-resolution satellite images were processed to complement the "QuickMap Services" and the formatting service of the Environmental Systems Research Institute. From the literature, 173 cenotes were identified as being sampled for various pollutants, and of these, one or more classes of pollutants were detected in 160 (i.e., greater than 92%) of the cenotes. Pollutants reported to be present included bacteria and viruses of human origin, fecal sterols, polycyclic aromatic hydrocarbons (PAHs), pesticides, pharmaceuticals, illicit drugs and personal care products. From the review of the literature, only 13 cenotes were reported to be free of the target pollutants. From this study, it can be concluded that the aquifer system with the Yucatan Peninsula is vulnerable to contamination from pollutants originating from wastewater, as well as surface runoff and infiltration from urban and agricultural lands.

The immense aquifer beneath the Yucatan Peninsula (YP), Mexico, is one of the least explored underground aquifer systems globally. The Great Mayan Aquifer (GMA) extends over more than 2000 square kilometers and contains what was recently recognized as the world's most extensive interconnected cave system: the 340-km-long Sac Actun/Dos Ojos system. On the YP, the aquifer is in direct contact with the surface through sinkholes formed by the collapse of caves in the karst bedrock (Gabriel et al. 2009). There are an estimated 9000 cenotes (from the Mayan word ts'óonot) on the YP (Lara-Lara et al. 2008), although the locations for most are imprecise due to difficult access (Moreno-Pérez et al. 2019). Ninety-nine are recognized as Ramsar sites, wetland designated to be of international importance, all located in the "Ring of the Cenotes" in the northern YP. The karstic limestone bedrock of the YP is highly porous, allowing rapid infiltration of surface precipitation and runoff pollutants (e.g., agrochemicals, livestock waste) into the aquifer (Moreno-Gómez et al. 2018). Pollution of interconnected ground and surface waters can occur by the intentional or accidental discharge of chemicals or waste into the environment. Pollutants such as fecal sterols, enteric bacteria, and persistent organic pollutants (POPs) have recently been documented in cenotes of the GMA. Of particular concern are the POPs, which are toxic chemicals that bio-accumulate in aquatic and terrestrial species, are environmentally persistent and can be transported long distances in the atmosphere. Among the POPs that have been detected in GMA cenotes are polycyclic aromatic hydrocarbons (PAHs) that originate

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mainly from anthropogenic activities such as vehicle emissions, petroleum products, and incomplete combustion of biomass. Another class of pollutants that has been found in the cenotes are organochlorine pesticides (OCPs) which are also persistent and subject to bioaccumulation, although many of these compounds have been banned for several decades. The presence of these chemicals in cenotes in the GMA highlights the vulnerability of the aquifer to environmental threats such as inadequate wastewater treatment, agricultural and livestock activities, and the rapid growth of urban areas and coastal tourism infrastructure across the YP. The objective of this investigation was to review the existing data on the type, location and distribution of pollutants in cenotes in the region, to identify the likely point and non-point sources and to recommend mitigation measures to reduce the threat of increasing levels of pollutants.

Methods

A systematic search was carried out in the PubMed and Web of Science databases of all scientific publications related to the terminology: "pollutants" "aquifer" "groundwater" "cenotes" "karst systems", and "sinkholes" of the Yucatan Peninsula. The cenote coordinate data gathered from the literature searches were linked to the geographic coordinates of the database of the Secretariat of Human Development and the Environment [SEDUMA] from 2018 and the Quintana Roo Speleological Survey [QRSS] of 2018, and these were transformed to geographic coordinates of the World Geodetic System [WGS84] of 1984 that allows locating any point on Earth with an error of fewer than two centimeters. A map was generated using the Geographic Information Systems Quantum (GISQ) version 3.1.2 geographic information system software. High-resolution satellite images were processed with the GISQ complement "QuickMap Services" and the formatting service of the Environmental Systems Research Institute (ESRI).

Results

Of the 1026 cenotes located within the YP, there are reports in the literature for 173 cenotes that have been examined for levels of pollutants. Of those studied, 160 cenotes (˃ 92%) were reported to be contaminated with detectable levels of one or more classes of contaminants, and only 13 cenotes (< 8%) were free of the targeted pollutants. The results of the survey of published data are summarized in Table 1 in chronological order. Microorganisms and sterols of fecal origin were the primary reported contaminants, being found in 112 of the evaluated cenotes (˂ 65%), while 14 of the evaluated cenotes contained PAHs (as many as 17 individual compounds) and OCPs were detected in more than 30 cenotes. The locations of these contaminated cenotes are illustrated in Fig. 1. The polluted cenotes on the satellite image showed them to be distributed across the entire YP, including in and near high-density populations, as well as in rural areas. Over thirty of these cenotes are located in Ramsar sites (Fig. 2). It must be noted that the classes of pollutants reported in these studies may reflect the analytical capabilities of laboratories in the region, as many laboratories do not have the advanced analytical instrumentation needed to detect some classes of contaminants, such as endocrine disrupting chemicals and current use pesticides.

Discussion

Microbiological Contamination

One of the main indicators of biological (microorganism) contamination in the water column and sediments of aquatic environments is the presence of sterols and enteric bacteria such as *E. coli* (Xie et al. 2017; Dayanti et al. 2018; Brisola et al. 2019). Fecal coliform concentrations found in cenotes (up to 8000 NMP/100 ml, fecal coliforms, 1–5207 μg/g dw total sterols) are at values higher than those recommended by the EPA (0 mg/L, public health goal). Urbanization, farming, population growth and tourism are significant factors that threaten groundwater quality (Moore et al. 2020; Li et al. 2021). The high microbial load in the aquifer is probably due to the State of Yucatan discharging over 102 M m^3 of wastewater into the aquifer without adequate treatment (CONAGUA 2012, 2014) and that as of 2000, there were more than 83,000 septic tanks in the state, most of which were not effectively treating household waste (Julia Pacheco et al. 2000). Easily transmitted by the fecal–oral contact, enteric pathogens in water can lead to adverse health effects (Dayanti et al. 2018). Some *E. coli* strains can also function as reservoirs of resistance genes that can be transferred to other pathogens through mobile elements and confer resistance to antibiotics such as beta-lactamases, fluoroquinolones, and quinolones (Jiang et al. 2011; Yeh et al. 2017). In a recent study by Moore et al. (2020), microorganisms showing antimicrobial resistance (AMR) were detected in several cenotes in the YP. In one study using phenotype and genotype biomarkers, *E. coli* strains isolated from water, soil and pig feces were highly resistant to sulfamethoxazole associated with colistin, enrofloxacin, and trimethoprim; there were even strains exhibiting resistance to multiple drugs (Brisola et al. 2019). One aggravating factor for fecal contamination in the GMA is that the YP is a major pig producing region, with over 194,000 tons produced Pork Meat in 2019 (SIAP 2019a). Many farms are small-scale and lack adequate animal waste treatment plants.

Table 1 Pollutants detected in cenotes of the Great Maya Aquifer on the Yucatan Peninsula

Cenotes evaluated	Cenotes polluted	Cenote name	Location	State	Assessed pollutant[a]	Concentration range[b]	Reference
3	3	X C U	N.r[c]	Yucatán Quintana Roo Yucatán	Enterobacteria *Escherichia coli* Enterococci, *Bacteroides* *Bifidobacterium*	N.r	Moore et al. (2020)
18	7	Encantado Tercer Cielo Dos Isos Dos Osos 1 Dos Ochos Cristal Escondido Juan Cenote Car Wash Tankah Cenote Tankah Road Gran Cenote Entrance Gran Cenote Exit Jailhouse Ak Tulum N-Tulum 2 Tankah Road W Jailhouse Cave Kaan Luun	Tulum	Quintana Roo	*Escherichia coli*	**0**–TNTC[d] CFU/100 mL[e]	Saint-Loup et al. (2018)
1	1	N.r.	Cancún	Quintana Roo	Naphthalene Phenanthrene	N.r	Gómez-Reyes et al. (2017)
8	8	N.r	Cancún Puerto Morelos	Quintana Roo	Total coliform *E. coli* PMMoV[f]	> 1–> 2420 MPN/100 mL[g] > 1–39 MPN/100 mL 1.7×10^{1}–1.0×10^{4} GC/L[h]	Rosiles-González et al. (2017)
13	13	Elepeten N.r Telchaquillo Nahyah Noh-Mozon Kalcuch Tanimax Yaxpakaltun Yaxcopoil X'batun San Ignacio N.r Doña Lucy	Dzilam de Bravo Cuzamá Celestun	Yucatán	Total sterols	> 1–> 5207 µg/g d.w.[i]	Derrien et al. (2015)

Table 1 (continued)

Cenotes evaluated	Cenotes polluted	Cenote name	Location	State	Assessed pollutant[a]	Concentration range[b]	Reference
48	48	N.r	Abala Cenotillo Chocholá Cuzamá Homún Huhí Izamal Kantunil Kaua Kopomá Mérida Motul Quintana Tizimín Tecoh Temozón Tinum Yaxcabá Valladolid	Yucatán	Fecal contamination	Positive result	Hoogestejin Ruel et al. (2015)
2	2	N.r	Cancún Playa del Carmen	Quintana Roo	Naphthalene Phenanthrene Benzo-α-anthracene Dibenzo (*α*,*β*) anthracene Indene Benzo-β-fluoranthene Benzo (k) Fluoranthene Phenanthrene	5.94 mg L^{-1} 0.90 mg L^{-1} 0.03 mg L^{-1} 0.82 mg L^{-1} 0.11 mg L^{-1} 0.07 mg L^{-1} 0.32 mg L^{-1} 1.54 mg L^{-1}	Lizardi-Jiménez et al. (2015)
20	20	Sabtun Xelactun Chunchucmil Chen Ha Yax Ha Kankirixche Sabak Ha Nayah X'pakay Uitzán Noria Tanimax Telchaquillo X'Kol- Ac Chen Vazquez Buenaventura Itzincab Dzonot Sabila Xlabon Dzonot Trejo	Celestun Kinchil Celestun Kopoma Chochola Abala Sacalum Tecoh Tekit Tekit Tecoh Tecoh Tecoh Izamal Buctzotz Buctzotz Buctzotz Dzilam González Dzilam González Dzilam González	Yucatán	α-endosulfan β-endosulfan Dieldrin 4,4'DDE[k] 4,4'DDD[l] Endrin Endrin aldehyde Endosulfan sulfate 4,4'DDT[m] Heptachlore α-lindane β-lindane γ-lindane δ-lindane	0.016–0.033 ppm[j] 0.014–0.027 ppm 0.003–0.059 ppm 0.081–0.112 ppm 0.017–0.109 ppm 0.248–0.359 ppm 0.068–0.120 ppm 0.024–0.036 ppm 0.016–0.032 ppm 0.421–2.804 ppm ND[n]—0.639 ppm ND—0.100 ppm 0.044—1.511 ppm ND—0.462 ppm	Polanco Rodríguez et al. (2015)

Table 1 (continued)

Cenotes evaluated	Cenotes polluted	Cenote name	Location	State	Assessed pollutant[a]	Concentration range[b]	Reference
28	28	N.r	Celestun Sisal Tetiz Hunucma Kinchil Samahil Tedzil San Antonio Mulix Cacao Abala Homun Cuzama Sabacche Poccheina Xcanchacan Dzonot -lu Dzilam González Dzilam de Bravo		Sterols Fecal sterols	0.5–˃2396 µg g^{-1} 0.3–˃1690 µg g^{-1}	Arcega-Cabrera et al. (2014)
8	8	Celestún Mono Xlaká Yal Ek Chan-Hulú Noh-hulú Alborada Sabak-há	Celestún Celestún Dzibilchaltún Abalá, Buctzotz Buctzotz Buctzotz Buctzotz	Yucatán	Aldrin Chlordane Hexachlorocyclohexane Endosulfan Endrin Heptachlor Metoxichlor DDT´s	ND—˃951 ng/L ND—˃1495 ng/L ND—˃2375 ng/L ND—˃1509 ng/L ND—˃1805 ng/L ND—˃1038 ng/L ND—˃935 ng/L ND—˃2355 ng/L	Cobos Gasca et al. (2014)
3	3	Alborada Yaal-Ek Noh-hulú	Celestún Abalá Buctzotz	Yucatán	Acetylcholinesterase activity as a biomarker for organophosphorus pesticides	N.r	Pacheco Garrio et al. (2014)
11	10	Talleres Rancho viejo R-510 Mojarras Siete bocas Xca–ha Chaac–mol Chanka–nab Bacalar Lagoon Milagros Lagoon Ojo de agua	Cancún Cancún Cancún Puerto Morelos Puerto Morelos Playa del Carmen Tulum Cozumel Chetumal Chetumal Holbox	Quintana Roo	Naphthalene Phenanthrene Hexadecane Naphthalene Pyrene Phenanthrene Hexadecane Phenanthrene Benzene Benzo (a) Pyrene Decane Hexadecane Naphthalene Naphthalene Hexadecane Naphthalene Pyrene No pollution	5.94 mg L^{-1} 0.07–0.09 mg L^{-1} 2.02 mg L^{-1} 0.12 mg L^{-1} 4.96 mg L^{-1} 0.53 mg L^{-1} 3.18 mg L^{-1} 2.54 mg L^{-1} 1.00 mg L^{-1} 9.67 mg L^{-1} 1.33 mg L^{-1} 5.87 mg L^{-1} 2.57 mg L^{-1} 3.48 mg L^{-1} 2.15 mg L^{-1} 2.18 mg L^{-1} 1.14 mg L^{-1} No pollution	Medina-Moreno et al. (2014)
1	1	Tank-ha	Tulum	Quintana Roo	Total coliforms *E. coli*	˃ 728 MPN/100 mL ˃ 186 MPN/100 mL	Leal-Bautista et al. (2013)

Table 1 (continued)

Cenotes evalu-ated	Cenotes polluted	Cenote name	Location	State	Assessed pollutant[a]	Concentration range[b]	Reference
2	2	N.r	Puerto Morelos	Quintana Roo	Total Coliform *E. coli* *Vibrio* spp. Caffeine	˃ 4–˃ 298 NMP/100 mL 0–85 NMP/100 mL Presence and Absence Presence and Absence	Leal-bautista et al. (2011)
2	1	Car Was N.r	Puerto Aven-turas Tulum	Quintana Roo	Phenanthrene Polycyclic musk Triclosan Nonylphenol Ibuprofen Naproxen Caffeine Cocaine Herbicides No pollution	2.12 ng/L 0.03 ng/L 0.81 ng/L 0.11 ng/L 4.27 ng/L 3.19 ng/L 12.50 ng/L 1.87 ng/L 2.39 ng/L No pollution	Metcalfe et al. (2011)
5	5	Sacbé Xcaret Xcaret Río Eden Calavera	Solidaridad	Quintana Roo	Fecal coliforms *Enterococcus* Total coliforms	0–386 NMP/100 mL 0–> 350 NMP/100 mL 0–> 24,196 NMP/100 mL	De la Lanza Espino et al. (2006)

All the pollutants reported are the only ones examined by the cited author. In no case, the presence of any pollutant was omitted, even if it was analyzed and undetected. In all cases, the concentrations of the various pollutants could vary according to the seasonal stage. In some cases, various cenotes could be evaluated by different researchers

[a]One or more contaminants found

[b]A concentration or ranges of concentrations found in 1 or more cenotes

[c]Not reported

[d]Too numerous to count

[e]Cologne Forming Unit/100 mL

[f]Occurrence of Pepper Mild Mottle Virus

[g]Most probable number/100 mL

[h]GC/L genome copies per liter

[i]dry weight

[j]Parts per million

[k]Dichlorodiphenyldichloroethylene

[l]Dichlorodiphenyldichloroethane

[m]Dichlorodiphenyltrichloroethane

[n]No detected

PAHs

One of the factors that cause the incidence of PAHs is the incomplete combustion of biomass. In the YP, it is local custom to set fire to agricultural fields each term of the productive cycle (known as nomadic or itinerant agriculture); this cultural characteristic could explain the presence of PAHs, since only in 2019 the planted area was greater than 12,800 ha (SIAP 2019b). PAHs found in cenotes, included high priority pollutants at concentrations above the EPA-recommended level of 0.0002 mg/L^{-1} (e.g., naphthalene at 5.94 mg/L^{-1}; pyrene at 4.96 mgL^{-1}; and phenanthrene at 1.54 mg/L^{-1}) (USEPA 2009). The hydrophobic nature of PAHs in aquatic environments allows them to adsorb to particles, leading to transport into sediments, where they are persistent (Aghadadashi et al. 2019; Asaoka et al. 2019; Stakėnienė et al. 2019). Humans can be exposed to PAHs in water via ingestion of contaminated drinking water or dermal contact (Thamaraiselvan et al. 2015; Hoseini et al. 2018; Ciarkowska et al. 2019). These compounds are known to be toxic to benthic organisms exposed to contaminated sediments and to terrestrial organisms exposed to contaminated soils (USEPA 2009). The potential for environmental effects caused by exposure to PAHs in cenotes in the GMA should be a research priority, since there have been few studies of the effects caused by the exposures to mixtures of PAHs (Ni et al. 2019). Studies carried out by Zhang et al. (2017) indicate

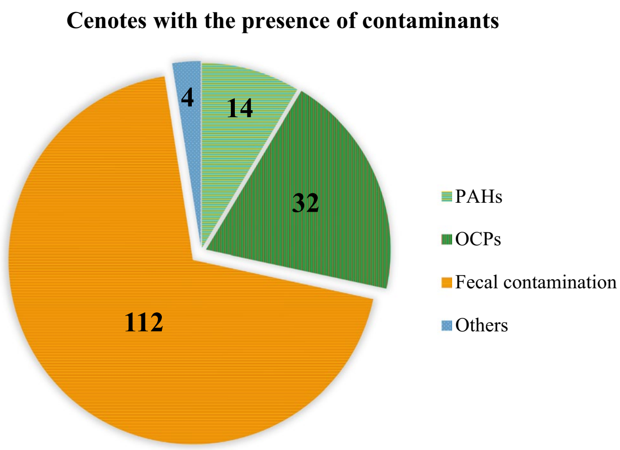

Fig. 1 Distribution of pollutants (one or more) found in cenotes of the Great Maya Aquifer on the Yucatan Peninsula

that PAHs deposited on impermeable surfaces in urban areas can be transported in rainwater to the aquifer, which would explain the distribution of these compounds in the aquifer in the YP region.

Pesticides

In Mexico, more than 47,000 tons (t) of active pesticide ingredients (> 29,000 t fungicides and bactericides; > 9000 t herbicides; and > 8000 t insecticides) were used (http://www.fao.org/faostat/en/#data/RP), reflecting a 65% increase in pesticide used over the last twenty years. Most of the OCPs detected in the studies reviewed here are now regulated under the control of the Stockholm Convention (UNEP 2005), of which Mexico is a signatory. According to this agreement use of aldrin, endrin and dieldrin are prohibited in Mexico. Nonetheless, controlled OCPs are still in use; for instance, there is no recorded use of heptachlor and HCH, but there is evidence of their presence. Moreover, after they were officially prohibited, documentation of chlordane, dichlorodiphenyltrichloroethane (DDT), lindane, and endosulfan was ended (SEMARNAT 2016). Nevertheless, concentrations of these and other compounds are still found that are far higher than EPA-recommended levels (α-endosulfan, 0.033 ppm; endrin, 0.359 ppm; dieldrin, 0.059 ppm; DDT, 0.036 ppm, and heptachlore, 2.80 ppm), (https://www.epa.gov/wqc/national-recommended-water-quality-criteria-human-health-criteria-table). The presence of OCPs such as

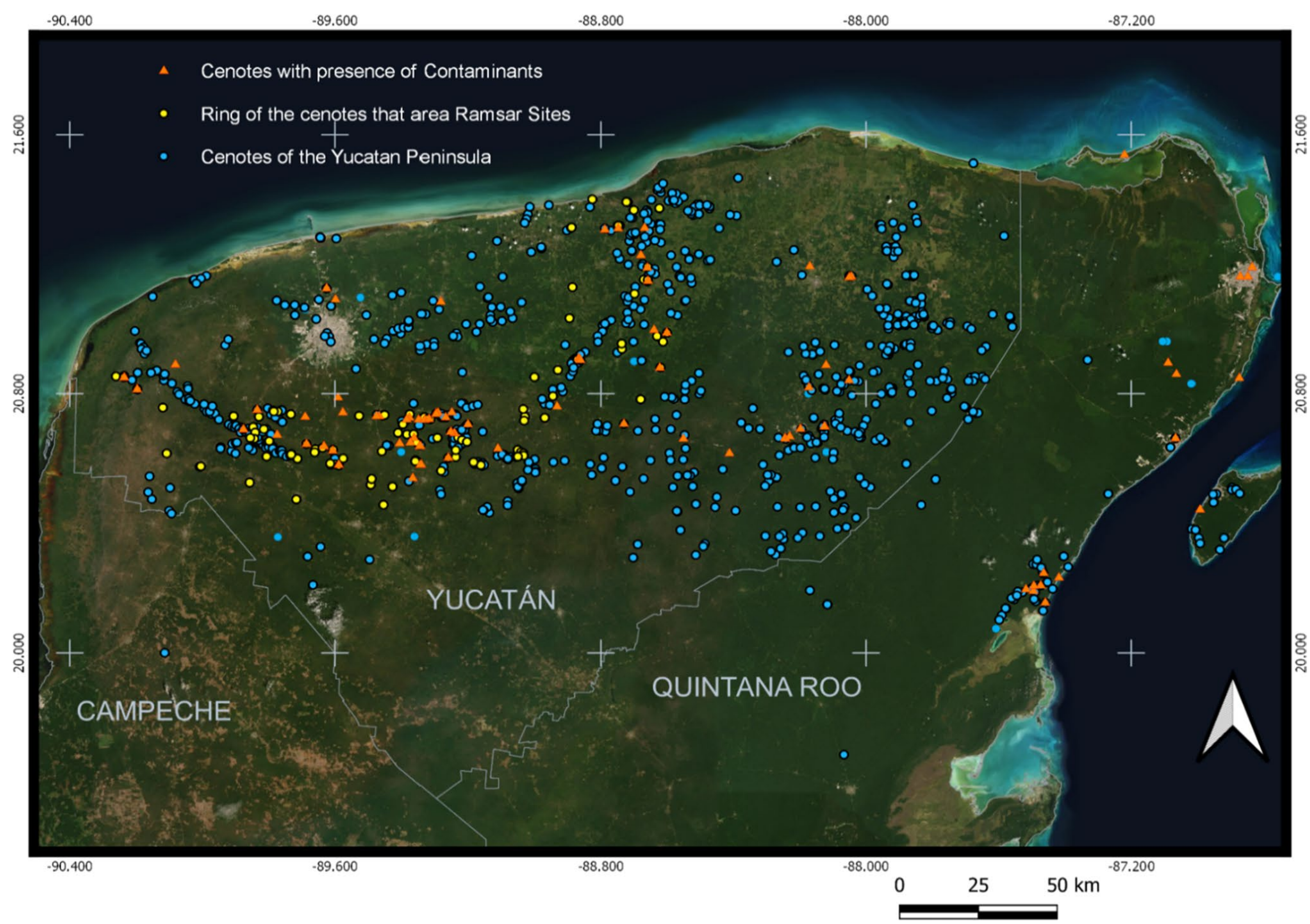

Fig. 2 Spatial distribution of georeferenced cenotes of the Great Maya Aquifer on the Yucatan Peninsula containing one or more pollutants

DDT in cenotes carries a severe environmental risk. Pacheco Garrido et al. (2014) detected reduced acetylcholinesterase activity in mosquito fish collected from cenotes in the GMA, which is an indicator of exposure to organophosphate insecticides. Metcalfe et al. (2011) detected several herbicides from the chlorophenoxy class in aquifers along the Caribbean coast of the YP.

Other Pollutants

Only one investigation of contaminants of emerging concern (CECs) was carried out in aquifers along the Caribbean coast of the YP, and several pharmaceuticals and personal care products were detected, including ibuprofen, naproxen, triclosan, and nonylphenol (Metcalfe et al. 2011). CECs are chemicals that have no regulatory standards, have recently been discovered in natural waters, can bioaccumulate in some cases, and create risks to human health, wildlife and the environment (USEPA 2017; Tang et al. 2019). Since these drugs are excreted by humans or make their way into sewage because they are used in a variety of cosmetics and other personal care products, they are indicators of contamination by domestic wastewater. The presence of these compounds in aquifers in the YP is consistent with reports of sterols, coliforms, *E. coli* and other microbiological organisms of fecal origin in cenotes in the region. Rosiles-Gonzales et al. (2017) evaluated the presence of Pepper mild mottle virus (PMMoV) one of the most abundant RNA viruses associated with human feces in various cenotes of the YP, finding its presence in all the cenotes evaluated.

The negative results obtained for the evaluation of pollutants existence in the 13 cenotes could be due to the evidence that they were not present. However, it is important to point out that different sampling sites, different layers and flow of water within the same cenote could affect the results; so there was not any evidence that cenotes in remote areas were less contaminated than cenotes located in tourist and/or urbanized areas.

Groundwater Vulnerability

Distribution maps of the sampled cenotes showed a large proportion of the GMA cenotes to contain various pollutants, including some cenotes near Ramsar sites. This situation can be a threat to the aquifer since the direction of groundwater flow is radial, starting in the southern part of the YP moving toward the coastal areas (INEGI 2002). This hydrological characteristic can favor the transport and distribution of pollutants within the entire aquifer. Worldwide, the number of classes of contaminants found in groundwater is increasing (Li et al. 2021); however, the interrelation of the various pollutants found in the YP cenotes and their possible synergistic effects on the environment is unclear. Although only 15% of the more than 1000 georeferenced cenotes (Moreno-Pérez et al. 2019) have been evaluated for pollutants, the results ($n = 160$ of 173) are representative. The overarching question is: if $^{>}$ 92% of the present samples exhibited some degree of pollution, how polluted is the GMA as a whole? The primary challenge to quantify its vulnerability is the very nature of this extensive underground aquifer system. A previous attempt to estimate groundwater vulnerability in the State of Yucatan was proposed by Moreno-Gómez et al. (2018). The authors used the European groundwater vulnerability method EPIK (i.e., Epikarst) that evaluates protective cover, infiltration conditions and development within the karst network, as well as a parameter indicating the effectiveness of the protective cover, depending on the thickness and hydraulic properties and the infiltration conditions. Another approach used was the COP method, which considers flow concentration (*C* factor), the properties of overlying layers above the water table (*O* factor), and precipitation (*P* factor) over the aquifer. Finally, the PaPRIKa method was used, which evaluates four criteria: *P* for protection of the soil cover, the unsaturated zone and the epikarst, *R* for rock type, *I* for infiltration and Ka for the degree of karstification. The authors found similarities in the methods used, indicating the state's groundwater to have a moderate vulnerability, but neither were congruent with regional characteristics, were not directly applicable and would need to be adapted to the region.

Addressing this imminent threat requires government willingness to promote efficient regulation of the prevention, detection and control of biological pollutants and POPs in aquatic environments. It is highly recommended to promote and invest in establishing an environmental mitigation industry focused on removing, managing, and degrading pollutants. Among the measures to mitigate the contamination of the GMA, it is necessary to identify the source and possible distribution of the pollutants to initiate confinement strategies and prevent infiltration into the aquifer. In addition, increasing the infrastructure and efficiency of wastewater treatment plants in urban areas and tourist centers and the design, construction and preventive maintenance of septic systems throughout the YP will reduce the pollution in the GMA.

In conclusions, more than 95% of the cenotes evaluated presented one or more pollutants. The distribution of the cenotes with the presence of pollutants is found throughout the YP. The cenotes called Ramsar sites are vulnerable due to the pollutants present in the aquifer; consequently, biological diversity, the environment, and human health are clearly at risk.

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Code Availability Not applicable.

Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Ethical Approval All authors are informed of the ethical guidelines of the journal and we declare that we have complied with them.

Consent to Participate All authors have participated in the design, planning and research of this work.

Consent for Publication All authors agree to the publication of this research according to the guidelines of the journal.

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