Dry, Salty, and Habitable: The Science of Alkaline Lakes

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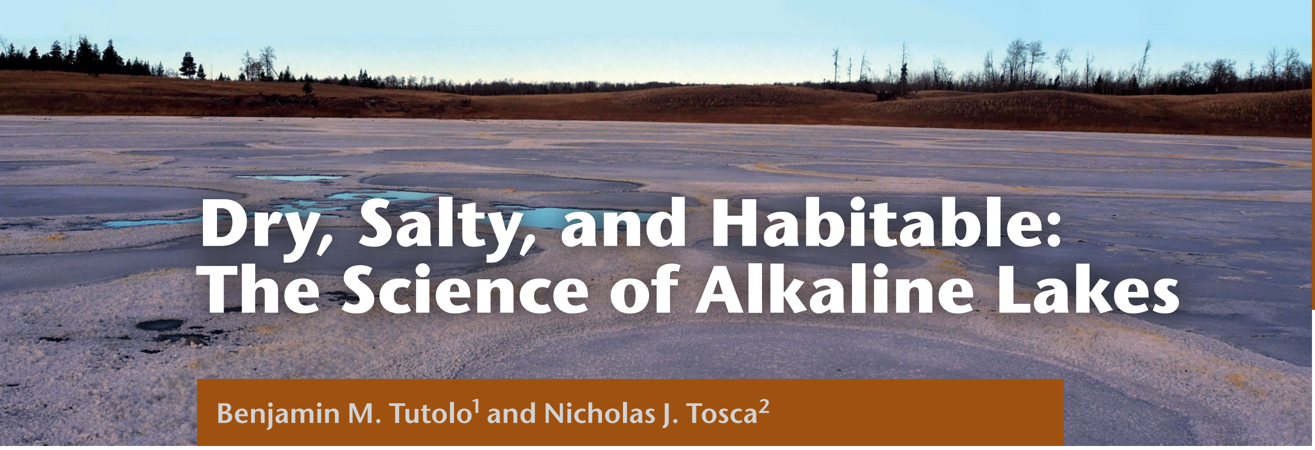

Alkaline Last Chance Lake, British Columbia, Canada, after complete evaporation. PHOTO: BENJAMIN TUTOLO.

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Alkaline lakes are incredibly dynamic, unique, and fascinating biogeochemical environments that have remained distinctive features of Earth's evolving surface over much of its history. Understanding these evaporative surface waters, their exceptionally productive ecosystems, and their rare sedimentary deposits requires an inherently interdisciplinary approach at the intersection of hydrology, geology, and biology. The discipline-spanning articles in this issue evaluate the diverse characteristics that make these dry, salty, and habitable environments so valuable in unraveling the history and evolution of Earth's surface, and in following the arc of habitability on ancient Mars. Here, in this introductory article, we summarize the characteristics and importance of alkaline lakes with the hope of attracting you, too, to join in our fascination with them.

KEYWORDS: alkaline lakes; biogeochemistry; Earth's resources; planetary habitability

INTRODUCTION

Alkaline lakes are standing water bodies with elevated pH, elevated carbonate alkalinity, or both. Though deceptively simple, achieving these defining characteristics requires a unique confluence of hydrologic, geochemical, and geomorphologic processes (FIG. 1). In a phrase, alkaline lakes are *dry, salty, and habitabl*e. To start, they occur in *dry* continental environments around the globe where evaporative fluxes outstrip hydrologic input for at least part of the year (FIG. 2). They become alkaline, rather than simply saline, during evaporation because of the unique chemistry of their inflowing waters (Tosca and Tutolo 2023a this issue). Secondly, extensive evaporation leaves their waters *salty*—many times saltier than even seawater, which is the saltiest naturally occurring water many of us will encounter over our lifetimes (FIG. 3). Together, these dry and salty conditions allow alkaline lakes to deposit uniquely identifiable, and economically important, sediments and sedimentary rocks (Raudsepp et al. 2023 this issue). Finally, because they bring together the ingredients, conditions, solvent, and access to energy required for life, alkaline lakes are uniquely *habitable* (Hoehler 2007). By virtue of their special chemistry, particularly their high carbonate alkalinity, and biological adaptations to it, alkaline lakes are among the most productive ecosystems on Earth's surface today (Haines et al. 2023 this issue), and are being intensively investigated as candidate environments that may have hosted the origins of life on Earth, and perhaps even Mars (Hurowitz et al. 2023 this issue).

Together, as the integrated products of hydrology, geology, and biology, alkaline lakes reflect distinctive characteristics of our own dynamic planet Earth. Like many features within the geological record, they have come and gone throughout Earth's history, ultimately contributing to our own societal advancement. Alkaline lakes have existed on Earth's surface for more than 2.5 billion years (Stüeken et al. 2015), and studies of ancient (older than around 3.5 billion years) sedimentary strata at Gale Crater, Mars, suggest that early Mars could have harbored alkaline lakes as well (Hurowitz et al. 2023 this issue). Over this time, a unique subset of Earth's organisms has adapted to thrive under alkaline lake conditions (Haines et al. 2023 this issue), in turn impacting the geochemistry and mineralogy of their sedimentary products (Chase et al. 2021; Raudsepp et al. 2023 this issue). Humans, too, have long made use of the unique benefits of alkaline lakes, in the case of ancient civilizations, by exploiting their salt deposits for processes as diverse as mummification, glass production, and nutrition (Raudsepp et al. 2023 this issue) and, in the case of

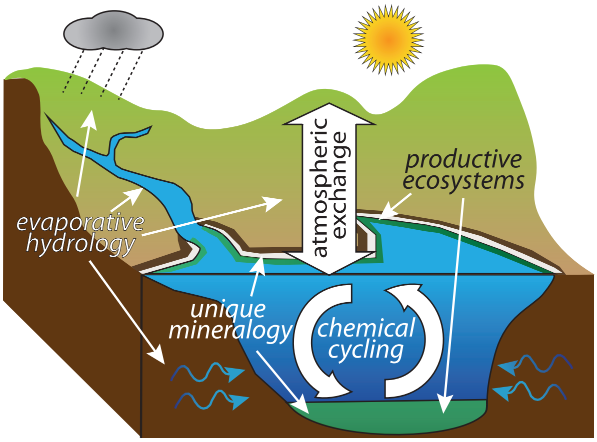

FIGURE 1 Efforts to understand the formation, evolution, geological preservation, and economic exploitation of alkaline lakes require an integrated approach that combines the geoscience subdisciplines of hydrology, mineralogy, geochemistry, and biology.

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modern society, by exploiting aspects of their biology and chemistry for processes as diverse as animal hide and paper processing (Haines et al. 2023 this issue). In the following, we introduce the science of alkaline lakes, as summarized in the articles included in this issue.

THE SCIENCE OF ALKALINE LAKES

Dry

Catchment hydrology is the primary environmental variable contributing to the genesis of saline lakes (Rosen 1994). Fundamentally, the process of evaporative concentration required to generate alkaline lakes (Tosca and Tutolo 2023a this issue) involves an unusual interplay between hydrologic inputs and outputs, namely, that lake water evaporation outstrips lake water supply (Fig. 1). Alkaline lakes typically form in basins or depressions whose hydrologic outflows are restricted by topographic or geologic features (Eugster and Hardie 1978). Nevertheless, to be classified as a lake, even an ephemeral one, these depressions must at least temporarily maintain standing water for part of the year. It is this combination of restricted basin conditions and outsized evaporative fluxes that contributes to the uniqueness and rarity of saline lakes, of which alkaline lakes constitute a key subset, as discussed further below.

The depressions that host alkaline lakes may be created through a variety of relatively widespread tectonic and geomorphologic processes. The commonality of these processes allows saline lakes to be widely distributed across the globe (Fig. 2). For example, tectonic extension and the coeval creation of topographic lows are likely factors in the production of some of the most well-studied active and ancient alkaline lakes, including the Cretaceous "presalt" stratigraphy of the South Atlantic (Saller et al. 2016), the Eocene Green River Formation (Lowenstein et al. 2017), and the modern East African Rift (Lowenstein et al. 2017). In other cases, the rain shadows that occur inland of mountainous continental margins (western North and South America) and inland plateaus (central Asia) yield prevalent, but often less expansive, saline lake systems (Eugster and Hardie 1978). Often, in this latter setting, the localized regions that host alkaline lakes are the expression of geomorphologic processes such as glacial retreat (Renaut and Long 1989). Given the often central role of plate tectonics in the generation of alkaline lakes on Earth, the question of whether alkaline lakes could have existed on early Mars in the absence of the processes that build mountains and rift continents on Earth requires viewing through a uniquely Martian lens (Hurowitz et al. 2023 this issue).

Sources of water to lakes include direct precipitation onto the lake surface, overland inflow (i.e., streams, springs, or runoff), and groundwater fluxes (Fig. 1). Each of these factors may be important to the generation, seasonal variation, and long-term fate of an alkaline lake. Yet, because the process of making an alkaline lake requires distinct, lithologically controlled chemistry (Tosca and Tutolo 2023a this issue), groundwater and/or surface water inflows must ultimately dominate the chemistry of hydrologic inputs to alkaline lakes. The closed or restricted nature of the basins characteristic of alkaline lakes suggests that the inflowing waters acquire this chemistry locally (Renaut and Long 1989), but there are exceptions such as lakes into which deep-cycled groundwater is the primary input (Rosen 1994; Lowenstein et al. 2017). Nevertheless, the sheer variety of saline lake chemistry in western North America (Eugster and Hardie 1978), which is made up of tectonic blocks of widely varying composition, suggests that catchment geology often dictates lacustrine chemistry.

Similar to input pathways, water removal from alkaline lakes occurs via a combination of groundwater seepage, overland outflow, and evaporation. Whether the dominant inputs to an alkaline lake are groundwater or surface water, the waters themselves are almost always dilute in nature. Thus, achieving the very significant increases in concentration required to produce alkaline lake waters (see Tosca and Tutolo 2023a this issue) and potentially produce significant deposits of economically and societally important minerals (Raudsepp et al. 2023 this issue), requires that evaporation is the dominant process by which water exits alkaline lakes. Observations of active alkaline lake systems, as well as the geologic record of alkaline lakes (Lowenstein et al. 2017), suggest that, while total desiccation often occurs, it is by no means a requirement. Indeed, Rosen (1994) points out that extensive evaporite accumulation can only occur under intermediate conditions, where evaporation is approximately balanced by water inputs. Ultimately, the unique hydrologic factors that either allow a lake to evaporate to dryness or to maintain long-lived, evaporite-depositing standing water must reflect an interplay between hydrogeology and climate. Reading the record of climatic change archived in alkaline lake deposits thus requires consideration of these hydrogeologic factors (Raudsepp et al. 2023 this issue).

To document the interplay between the abundance of alkaline lakes and climate, we need look no further than anthropogenic climate change as a driver of evolution of lake systems. Observed and predicted climate changes suggest that the abundance of underfilled lakes has been increasing and will continue to do so as anthropogenic CO_2 emissions raise global temperatures (Wang et al. 2018). Indeed, these stark predictions imply aridification in locations where saline lakes exist today (Fig. 2), as well as increases in evaporation from lakes in locations with few such lakes (Wang et al. 2018). This, in turn, suggests that periods of climatic change would have led to simultaneous changes in the abundance of alkaline lakes over Earth's history. Inasmuch as drying conditions are required to facilitate prebiotic reactions in lacustrine environments, periods of global drying on early Earth and Mars may have

Figure 2 Global distribution of saline lakes (after Williams 1996). Lakes in these regions with the appropriate inflowing chemistry will develop into alkaline lakes upon evaporative concentration (see Tosca and Tutolo 2023a this issue).

facilitated the reactions leading to life's origins (Hurowitz et al. 2023 this issue). In this way, Darwin's "warm little pond" proposition for the origin of life on Earth may have been more prescient than even Darwin himself could have guessed.

Salty

While hydrology dictates the genesis of saline lakes, it is the chemistry of the inflowing waters that controls whether they become alkaline, or simply saline, upon evaporation. The fundamental chemical requirement for the generation of an alkaline lake is that the concentration of carbonate alkalinity must be greater than the concentration of Ca^{2+} in the inflowing water (Tosca and Tutolo 2023a this issue). This requirement, and the charge balance constraints imposed by alkalinity relationships in aqueous solutions open to the atmosphere (Tosca and Tutolo 2023b, *Toolkit*, this issue), demands that alkaline lake source waters must have interacted with rocks that more readily donate ions such as Na^+ and K^+ than Ca^{2+}, a fact that offers unique insight into the catchment lithologies capable of generating alkaline lakes. Modern and ancient alkaline lakes, such as those of the Cariboo Plateau in Canada (Renaut and Long 1989) and the ancient "presalt" lakes of the South Atlantic coastal margins (Saller et al. 2016), often co-occur with basaltic lithologies. Yet, it seems that evaporative lakes hosted entirely within basaltic catchments are unlikely to develop alkaline chemistry without other, more petrologically evolved lithological influences (Renaut and Long 1989; Tutolo and Tosca 2018), again creating unique challenges for studying the presence and characteristics of alkaline lakes on the largely basaltic surface of Mars (Hurowitz et al. 2023 this issue).

Waters with the required carbonate alkalinity to Ca^{2+} ratio (>>2) invariably produce unique sedimentary deposits upon evaporation. These sediments are characterized by a startling diversity of minerals, and efforts to study them all but require a crash course in unfamiliar sulfates, borates, and carbonates (Raudsepp et al. 2023 this issue). The distinct chemistry of these minerals, and their silicate and halide companions, reflects not only the composition of the inflowing waters but also the environmental conditions under which they were precipitated (Fig. 3). While Ca and Mg carbonates are common products of alkaline lake sedimentation (Tutolo and Tosca 2018; Raudsepp et al. 2023 this issue), the extraordinarily high alkalinity of these lake waters (often 100s or 1000s of times more concentrated than that of seawater) dictates that they contain little Ca or Mg, because of the low solubility of Ca and Mg carbonates in these waters. Rather, Ca and/or Mg carbonate mineral growth during the initial stages of evaporation is key to the eventual development of alkaline lakes (Tosca and Tutolo 2023a this issue). These unique conditions lead to unusual ("nonclassical") carbonate nucleation and growth pathways (Raudsepp et al. 2023 this issue) that invite special consideration of often unconsidered growth inhibitors, such as phosphate (Pietzsch et al. 2022). In fact, the elevated carbonate concentrations and synchronously low Ca concentrations in alkaline lake waters allow phosphate to behave essentially conservatively during evaporation as a result of the elimination of the apatite mineral control on phosphate concentrations characteristic of most other natural waters (Toner and Catling 2020), which, in turn, drives hypotheses for alkaline lakes as loci for prebiotic chemistry (Hurowitz et al. 2023 this issue).

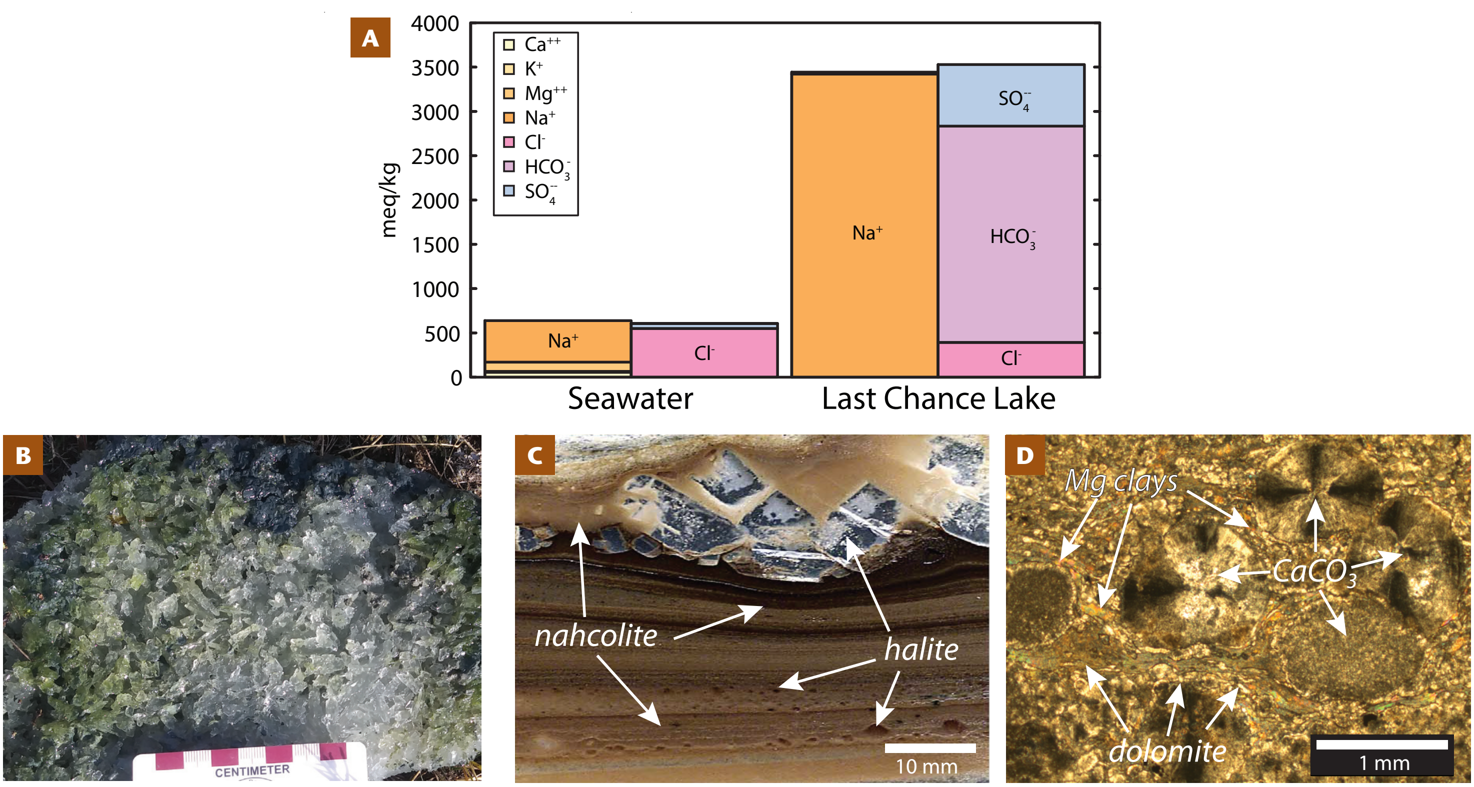

Figure 3 Examples of the unique geochemistry and mineralogy of alkaline lakes. (**A**) Representative ion balance of Last Chance Lake, British Columbia (Renaut and Long 1989), compared with the ion balance of seawater. (**B**) Interlocking trona ($Na_3(HCO_3)(CO_3)\cdot 2H_2O$) crystals recovered from the subsurface of Last Chance Lake. Photo: B. Tutolo. (**C**) Thin section micrograph of the Eocene Green River Formation, showing layered nahcolite ($NaHCO_3$) and two forms of halite (NaCl): smaller rafts (formed on the ancient lake surface) and larger cubes (formed on the lake bottom) (after Lowenstein et al. 2017). (**D**) Thin section micrograph of the Cretaceous Barra Velha Formation of the Santos Basin, offshore Brazil, showing magnesian clays (e.g., kerolite ($Mg_3Si_4O_{10}(OH)_2\cdot nH_2O$), calcite ($CaCO_3$), dolomite ($CaMg(CO_3)_2$) (after Tutolo and Tosca 2018).

Habitable

Together, the hydrology and geochemistry of alkaline lakes allow them to become uniquely productive geologic environments that, in turn, also contribute to planetary habitability. Although they may be volumetrically minor compared with other, fresher lakes and the oceans, alkaline lakes contain much, much more carbon per kilogram than circumneutral pH surface waters like seawater (Fig. 3) and thus play a major role in regulating Earth's global carbon cycle and its coupled effects on climate (Cole et al. 2007; Tranvik et al. 2009). Carbon dioxide from the atmosphere or volcanic or metamorphic outgassing is absorbed into lake waters (Edmonds et al. 2020) and partially converted into organic matter by their resident biota. This organic matter may either be preserved through sedimentation and burial (e.g., Saller et al. 2016) or re-oxidized to CO_2 and returned to the atmosphere (Haines et al. 2023 this issue). If sedimentation rates are high and the organic matter is rapidly buried, an alkaline lake is able to take a larger bite out of the global carbon budget than if sedimentation is slow and the organic matter is rapidly oxidized back to CO_2. The dynamics of carbon burial in alkaline lakes are ultimately controlled by a wide range of factors, from lake depth and redox stratification to hydrologic inputs.

The challenging geochemistry of alkaline lake waters requires that their inhabitants acquire unique evolutionary adaptations to exploit the inorganic carbon wealth available to them (Haines et al. 2023 this issue). Alkaline lake waters have much in common with industrial process waters used to prepare leather, whiten paper, and launder fabrics, such that these industries have unearthed environmentally friendly alternatives via the selective manipulation of the evolutionary advantages of alkaline lake organisms (Haines et al. 2023 this issue). Incredibly, alkaline lake organisms have long provided spirulina, a widely marketed, sustainable source of protein (Haines et al. 2023 this issue).

If hydrologic and geochemical factors converge to create alkaline lakes on prebiotic planetary surfaces, their coupled, elevated phosphorus concentrations may provide the vital kick needed to facilitate organic synthesis reactions (Toner and Catling 2020). In fact, theoretical calculations indicate that prebiotic alkaline lakes may have also concentrated cyanide, which, while toxic to many modern organisms, is a critical feedstock molecule required for biomolecular synthesis, in the form of highly soluble ferrocyanide salts (Toner and Catling 2019). These chemical features, in combination with access to ultraviolet light, and the influence of dynamic physical and chemical cycling, have motivated the intense study of alkaline lakes as potential environments that may have hosted the chemical origins of life on Earth (Toner and Catling 2019, 2020; Hurowitz et al. 2023 this issue). An immediate question, of course, is the degree to which these advantages extend to other rocky planetary surfaces, and if so, how frequently such environmental requirements may have been fulfilled (Hurowitz et al. 2023 this issue). For the first time, observational constraints from Mars exploration missions mean that an initial explanation is potentially within our grasp. Available data indicate that lakes associated with high carbonate alkalinity do not appear to have been widespread on the surface of Mars, but alkaline waters may have been more commonly generated at the interface between ancient surface and subsurface hydrological systems (Hurowitz et al. 2023 this issue). Remarkably, as these words are being written, the *Perseverance* rover is collecting geochemical and mineralogical data, as well as the samples from which they were derived, that are likely to test these hypotheses. Ultimately, these samples will be returned to Earth (and perhaps made accessible to some of the younger readers of this issue) in the coming decade.

HISTORY AND OUTLOOK

As with so many geologic phenomena, the history of alkaline lakes is likely similar in length to the history of our planet. Likewise, the history of human attraction to these distinctive environments in all probability dates back to the dawn of human civilization. As reviewed by Raudsepp et al. (2023 this issue), the ancient Egyptians and Romans were exploiting alkaline lake deposits to provide material for the production of glass, bread, and medicine as early as 3500 BCE (Kostick 1983). To this end, much of our understanding of alkaline lakes has in fact been driven by their societal value. Although they were likely well known to the indigenous peoples who had populated western North America for millennia, some of the earliest, published scientific research on saline and alkaline lakes in the young country of Canada (confederated in 1867) was performed to quantify the reserves of valuable mineral deposits in the region (Goudge 1926) and to understand the developed methods for extraction (Jenkins 1918). Any scientist reading this article who has had the pleasure of visiting these lake systems will quickly realize how difficult the path to scientific discovery in these settings must have been for those early researchers. Indeed, as reported by Jenkins (1918), "beneath this white film [of efflorescent salts] is mud, black, foul, and treacherous, which has been the cause of the miring of cattle in the past." Jenkins (1918) does not mention insects, but interested readers can take our word that those looking for the products of the most productive ecosystems on the planet (Haines et al. 2023 this issue) will certainly not be disappointed by the biomass of mosquitos produced during the summer on the Cariboo Plateau. Thus, the history of the science of alkaline lakes is not just one of perseverant scientists seeking to understand, develop, and exploit products of critical societal value but also one of the development of high-performance materials and chemicals that allow humans to maintain a (semi-) comfortable existence while exploring them.

Building on this lengthy history, the articles in this issue of *Elements* review the remarkable scientific and societal significance of alkaline lakes. Consistent with this magazine's broad readership and diverse coalition of allied societies, these articles delve into the knowledge and tools used and created by the wide-ranging teams of scientists studying these unique water bodies and their sedimentary deposits. A collection of five magazine-length articles could never encompass the exceptional breadth of information associated with this field of study, but we nevertheless hope that each reader will find something to like, and something that they will hope to learn more about.

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