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Ideas and Perspectives:

Value and efficiency of ocean sample return and laboratory analysis: Key analytes are orders of magnitude rarer than a needle in a haystack

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Abstract

There is a pressing need to understand how the oceans are responding to human perturbations, yet their vastness poses a considerable challenge. The development and deployment of biologically and ecologically relevant in situ sensors are frequently invoked as a means to improve this understanding; however, there are still relatively few sensors for these parameters due to the analytical challenges involved in their measurement. Specifically, many analytes of interest are orders of magnitude rarer by mass than a needle in a haystack (e.g., dissolved iron and eDNA are 10^4 to 10^8 times rarer), and this scarcity creates challenges involving preconcentration, interfering elements or compounds, and analytical methods difficult to perform underwater. While sensor development is important, the role of sample collection and return is also an important, yet oft overlooked, complement for studying ocean change and is ripe for additional development. Laboratory analyses of returned samples can be high throughput for challenging analytes like micronutrients and omics parameters, and these techniques are operational now. Moreover, sample return allows parallel analyses and archiving through splitting of sample material. Similar to human health diagnostics, laboratory analyses can provide ocean health diagnostic capabilities showing how ecosystems are responding, complementing sensor-based vital sign measurements indicating environmental change. Together, these properties make sample collection and return a critical component for achieving the global ocean coverage needed to capture the environmental changes well underway today, and this approach should continue to be factored into ocean observing strategies.

Introduction: Is a goal of robust sensors for all ocean parameters feasible?

Throughout the history of ocean science, our understanding has been shaped by a combination of observations made in situ (underwater) or by discrete samples that are analyzed at sea or returned to home laboratories. Ship-based sampling can include a variety of wire-based deployments (e.g., CTDs) and flow-through seawater systems, and other platforms take many forms including coastal monitoring stations, floating moorings tethered to the seafloor, and remotely operated and autonomous robotic vehicles. With the emergence of underwater and autonomous platforms there has been keen interest in the development of robust in situ sensors for key environmental parameters. By robust sensors we refer to devices that can routinely be deployed underwater including those that can be remotely deployed for months to years without maintenance or replenishment of reagents. Due to this interest, there has been an implicit, and

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more recently explicit, expectation that the development of new robust sensors for any and all parameters of interest is both imminent and inevitable.

Yet the emergence of reliable low-cost sensors for the majority of biological and chemical parameters has not matched the pace of platform development. Why have expectations and reality diverged? And what are the resulting implications when Earth observations are critical in a time of rapid environmental change? Here we discuss the disconnect between the assumption of 'imminent and inevitable' biological and chemical sensor development with the reality of analytical chemistry challenges, and we make the case for appreciating the value of 'less exciting' sample return strategies in ocean research programs.

The challenges of in situ analysis for scarce and complex analytes

One can understand how the aspirational idea that any property of interest should be measurable by a sensor might arise. The robustness of now-ubiquitous oceanographic sensors for temperature, pressure, salinity, oxygen, and fluorescence has transformed the field of oceanography over recent decades. Their impact is demonstrated by the fact that the main shipboard instrument, the CTD water sampling rosette, is commonly referred to simply by the abbreviation of its three key sensors: *c*onductivity (from which salinity is derived), *t*emperature, and *d*epth (derived from pressure): the "CTD". The sensors on rosettes operate without reagents, can operate to full ocean depths, and are robust for months to years, only needing periodic recalibrations.

There have been important efforts to develop in situ analytical systems for parameters beyond these examples, including complex biological and chemical parameters (Aguiar et al., 2023; Grand et al., 2016; Geißler et al., 2021; Hurst et al., 2010; Johnson and Coletti, 2002; Ma et al., 2016; Pierrot et al., 2009; Vuillemin et al., 2009; Wang et al., 2015; Zeng et al., 2003). Notably, there are also instances of new automated shipboard systems for analytes such as inorganic carbonate parameters (e.g., pCO_2) and flow cytometers for microbial cell abundances (in situ and shipboard: Olson et al., 2003; Swalwell et al., 2011). Finally, there are also capabilities for mooring and AUV-based in situ DNA analysis using DNA-specific probes and quantitative polymerase chain reaction methodology (qPCR; Scholin et al., 2009; Scholin et al., 2017; Yamahara et al., 2019). These are some recent examples of technological implementations of difficult analytical measurements underwater or at sea (e.g., shipboard DNA sequencing). These technologies are capable of conducting in situ analyses with high temporal resolution and

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provide valuable new research insights and public health information (e.g., harmful algal bloom dynamics). Yet in most cases, these newer systems also require complex instrumentation and/or perishable reagents and hence are usually distinct from CTD sensors that are robust for long deployments. Due to reagent replenishment, instrument durability, power needs, or cost, many of these devices may not yet be compatible with being integrated into long-term oceanographic systems such as floats and offshore moorings or with application at global scales. And in many cases, these instruments are in fact collecting and processing samples in situ: performing separation, chemical preprocessing, analysis, and archival storage, with the advantage of gaining the final data sooner than returning the sample for shore-based analysis. Yet, there are trade-offs in utilizing in situ analysis, where results are often more limited in scientific scope, generally focusing on a single parameter or method per instrument, compared to what is possible by applying multi-analyte laboratory workflows to returned samples. For example, in situ qPCR only detects specific predetermined DNA sequence targets, limiting analysis to targets chosen prior to the assay. In contrast, widely used sequencing-based approaches such as metabarcoding, metagenomics, and metatranscriptomics, which operate on returned samples, can broadly survey the nucleic acid assemblage.

Given the rapid pace of technological development, one might assume an inevitable expansion of the CTD to a “CTD-CNPFDPMV” system (carbon, nitrogen, phosphate, iron, DNA, protein, metabolites, vitamins, etc.). With a few exceptions, this has largely not happened (Johnson et al., 2017a; Johnson et al., 2017b; Wang et al., 2019). The nitrate sensor is one example of successful sensor development of a key biogeochemical parameter that overcame potential interference from bromide (Johnson and Coletti, 2002), yet still faces detection limit challenges in low-level oceanic environments. The promise of a broad array of robust and sensitive biological and chemical sensors that replace laboratory capabilities remains largely unfulfilled. Why? Aside from the unique engineering challenges that confront measurements in the ocean environment, there are fundamental chemical challenges to the development of sensors. Primary among these is the low concentration of many analytes of interest within a complex matrix of interferences (Table S2).

To demonstrate this, we can look at the salinity sensor, which is considered by many to be an ideal chemical sensor of seawater given its durability and precision and thus one that other chemical sensors might aim to replicate in its simplicity. It is in fact an electrical sensor that measures aqueous conductivity generated by the abundance and activities of major cations and anions in seawater, in particular sodium and chloride, as well as some lesser contributors

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(sulfate, potassium, magnesium, and calcium)(Jacobson, 1950; Gregg and Cox, 1971; Gregg and Cox, 1972). The abundances of sodium and chloride are very high at ~0.5 mol L^{-1} each, and the other major salts are approximately tenfold lower than that. Together these charged ions create an electrical conductivity that solid-state sensors can measure precisely, and from which salinity is then calculated. Oceanographic processes that vary the salinity content in seawater, such as freshwater inputs and evaporation, cause variations in seawater salinity that are imprinted onto the long timescales of ocean mixing. While salinity is an essential parameter in the study of ocean circulation, the variations in salt chemical composition are not themselves major influences on ocean biogeochemical processes, with the ratio of major salts being a conservative property that is relatively invariant throughout much of the ocean. It is worth mentioning that even for this ubiquitous measurement, laboratory-based instrument calibrations are still necessary to detect small changes in salinity attributed to climate change (Castagno et al., 2019).

In contrast to salinity's role in tracing physical processes, the nutrients and micronutrients in seawater have a major influence on ocean biogeochemical processes, including the marine carbon cycle, and hence are desired biogeochemical analytes. For example, iron directly controls the photosynthetic productivity of a third of the surface ocean, and up to 70% of productivity when its limitation of nitrogen fixation is included (Moore et al., 2004). Its dissolved concentration ranges are often sub-nanomolar, for example as low as 0.15 ± 0.08 nM (n = 141) in the Pacific Sector of Antarctica, an iron-limited region (Tagliabue et al., 2012). This is more than nine orders of magnitude lower in abundance than the co-occurring major salts sodium and chloride.

For comparison, we can utilize the English expression "like finding a needle in a haystack", which was already considered an "old adage" in the 18th century (Rogers, 1779). The expression refers to something that is near impossible to locate. Using the average weight of modern metal hand-sewing needles and a literature estimate of a haystack's mass, we can estimate that the mass of a needle in a haystack is actually ~13,000 times more abundant than the dissolved iron in seawater (also by mass; Figures 1 and 2 and Table S1; Supplemental Methods). Many key vitamins, metabolites, micronutrients, and biomolecules (specific nucleic acid or protein sequences) can be similarly scarce or even scarcer. For example, eDNA, referring to environmental DNA including remnants of larger passing organisms (e.g., small animals and fish), can provide taxon-specific sequences that are rare despite the overall aggregate (primarily microbial) DNA being more abundant in seawater. In this analogy, salinity

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sensors are measuring the haystack itself: the salts that provide the signal are the major chemical constituents of seawater after water itself, contributing ~3% of the weight of seawater. Finding a steel needle in a haystack might be easier today, as metal needles have ferromagnetic properties allowing electromagnetic approaches to be employed.

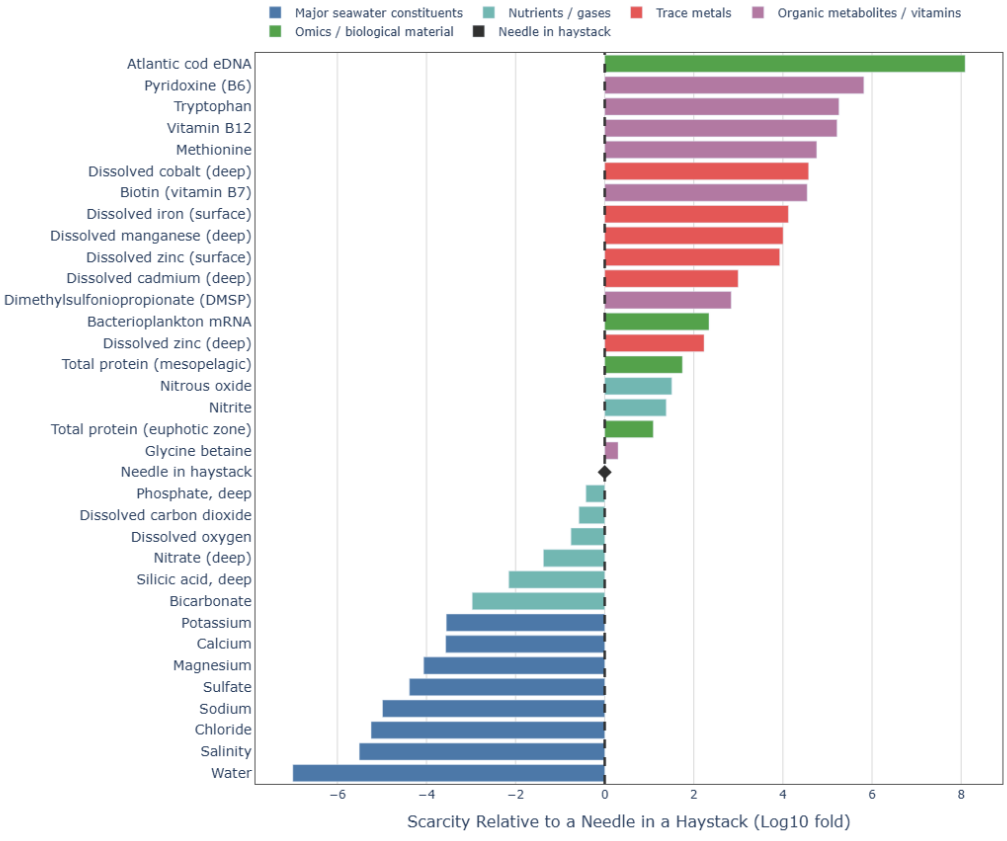

Figure 1. The scarcity of example analytes in seawater, presented normalized to the challenge of finding a needle in a haystack (mass-to-mass basis). Analyte list is not exhaustive. See Table S2 and Supplementary Material for information on calculations and references.

Despite these 13,000-times-harder-than-a-needle-in-a-haystack challenges, marine chemists have excelled at measuring iron and other trace metals in seawater thanks to the development of trace metal clean sampling and analytical methods and extensive intercalibration efforts like those of the GEOTRACES program. In recent years, GEOTRACES has mapped the distribution of trace elements on a global scale for the first time (Schlitzer et al., 2018; Anderson et al.,

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2020). The methods used include flow injection, atomic absorption, electrochemistry, and inductively coupled plasma mass spectrometry approaches. These are methods requiring laboratory expertise, sophisticated instrumentation, and specialized reagents. Some such as flow injection (adapted from microfluidics) and electrochemistry are capable of deployment as in situ analytical systems (Achterberg et al., 1999; Geißler et al., 2017), but generally lack the ability to analyze metals complexed to organic molecules or to filter in situ, and hence capture a subset of the metal inventory compared with ship-based or shore-based methods that can differentiate between dissolved, particulate, labile and complexed, and colloidal fractions of the total metals found in seawater. Moreover, the stability of chemical or enzymatic reagents continues to be a significant challenge for longer underwater deployments. It is also important to recognize that most in situ chemical sensors are single analyte: individual systems would have to be designed and deployed for each parameter (e.g., iron and zinc sensors would have to be separate), rather than as multi-analyte mass spectrometry methods commonly used with samples returned to the laboratory (Jackson et al., 2018). Many analytical methods are also affected by interferences from co-occurring elements. Perhaps because of these challenges, in situ seawater metal analyses are currently rarely used compared to ship-based or land-based laboratory methods.

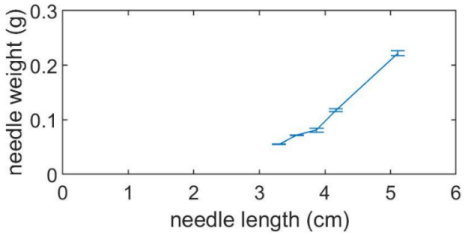

Figure 2. Weight of sewing needles relative to length. Relative standard deviation (RSD) of weights varied from 0.9 to 4.9%. The needle weight used in Figure 1 was the mean across five size classes, reported as 0.1095 ± 0.0020 g (see Supplementary Materials and Table S1).

Similarly, there are numerous biological molecules that are of interest to oceanographers. Chlorophyll and colored dissolved organic matter (CDOM) are highly abundant and have optical properties that allow them to be measured by robust sensors, yet many biomolecules in seawater either don't share these optical properties or aren't as abundant. DNA, RNA, proteins, and metabolites (where a collective 'omics' term refers to deep molecular methods including

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genomics, transcriptomics, proteomics, and metabolomics that measure the respective biomolecules) are of great interest to researchers for their ability to understand the mechanistic workings of organisms and ecosystems. Instruments for in situ sensing of DNA have utilized PCR technology for specific sequences such as those of red tide harmful algal blooms – when rapid results regarding a known target can be informative for human health issues (Scholin et al., 2009). But other omics analyses require ship- or shore-based laboratory processing, for example metabarcoding (amplicon sequencing) and the targets of these analyses are not necessarily known in advance (e.g., the analyses may be exploratory). Metabarcoding can capture a wide range of taxonomic targets ranging from microbes to macro and megafauna, and the concentrations and distributions of their associated nucleic acids may be taxon-specific, making a "one-size-fits-all" sensor unrealistic. For example, omics data on microbial targets (where the processed molecules may stem from the organisms themselves) may only require small collection volumes (Patin and Goodwin, 2023), but animal target detection (based on fragments shed by organisms of various sizes, DNA shedding properties, population densities, and distributions) may require much larger volumes (Govindarajan et al., 2022; Herrera et al., 2026). Similarly, for measurement of proteins there is the potential for adopting antigen assays for specific proteins, similar to those used in over-the-counter COVID-19 test kits, but significant investment and testing is needed for each protein target, and many such assays are needed to capture ocean diversity making the progression to deployment challenging. While these in situ approaches are technically feasible, they can involve complex instrumentation and regular upkeep, and often sample a narrow range of biological diversity. Hence these approaches are often well-suited as key components of coastal monitoring systems or research on specific questions rather than for global oceanographic diversity and functional biogeochemical studies.

The Emergence and Power of Multi-Analyte Laboratory Analyses

In contrast to the current state of in situ capabilities described above, laboratory capabilities today far exceed what can be done underwater. There is incredible scientific depth in current chemical and biomolecular measurements that can be conducted on samples returned to the laboratory. Samples can be analyzed for many parameters (dozens of elements) or diverse molecules (hundreds to many tens of thousands or more) within broader parameter classes (i.e., eDNA, eRNA, proteins, metabolites) simultaneously from splits on each individual sample, including analytes that were not originally planned (e.g., Gold et al., 2023). Moreover, samples can be archived for future analyses that can be used to answer new questions or to apply new approaches, including assays still under development. Publicly accessible archives and data

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repositories can also make samples and data available to scientists who do not have the resources for their own seagoing expeditions.

In recent decades, the capabilities of laboratory technology have become exceptional. Mass spectrometers, nucleotide sequencers, and other instruments are capable of highly sensitive and information-rich analysis of atoms and molecules within complex samples. Mass spectrometers are cornerstones of both inorganic and organic analyses, where they separate molecules or elements from interferences, allowing sensitive and accurate measurements. Improvements in mass spectrometers have also enabled many analytes (hundreds to hundreds of thousands) to be measured on a single sample injection, providing great efficiency and depth to the analyses. While simple field mass spectrometers for dissolved gases exist, most mass spectrometers have attributes that are difficult to adapt and maintain in underwater (and shipboard) environments such as high vacuum and power requirements, gas and solvent use, complex electronics, and sophisticated inlet systems. Similarly, nucleotide sequencing has increased its depth of analysis with successive generations of technology, now providing millions of reads per sample. There are domain cultural differences here that we see play out in rare cross-disciplinary conversations: oceanographers accustomed to a handful of individual sensors providing tens of thousands of data points per cast might consider a vertical profile with 20 discrete samples to be greatly lacking. Whereas scientists now generate omics data with millions of valuable data units (base pairs or spectral features) from every discrete bottle or pump depth, each providing a wealth of biological and chemical information.

The potential for omic data to contribute diagnostic information to understanding ocean health parallels activities in biomedical fields. The discovery of biomarkers that can diagnose various cancer types based on blood sample analysis has been relatively challenging despite great investment (Hristova et al., 2019; Ren et al., 2020). In contrast, the discovery of biomarkers for nutritional controls on ocean productivity has been rapid and the results robust, exemplifying a valuable contribution of the omics revolution to marine sciences (Chappell et al., 2012; Cohen et al., 2021; Frischkorn et al., 2019; Held et al., 2020; Kellogg et al., 2022; Saito et al., 2014; Saito et al., 2015; Ustick et al., 2021). Biomarkers, represented by proteins, transcripts, or genes, that aid in the acquisition or sparing of key nutrients have been discovered in marine phytoplankton for nitrogen, phosphorus, iron, zinc, and vitamin B_{12} (Bertrand et al., 2012; Walworth et al., 2022; Kellogg et al., 2022). Moreover, because of the sequence specificity of the genes and proteins being measured, it is possible to discern who is experiencing nutritional stress to precise taxonomic levels (genus, species, or even ecotype level; Saito et al., 2015, Yang et al., 2026).

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In short, within a single sample, omics measurements can discern who is present, what is controlling their productivity (e.g., their carbon fixation rates), and even make estimates of their biogeochemical reaction rates (Saito et al., 2020).

Figure 3. The Autonomous Underwater Vehicle Clio, capable of high-throughput full ocean depth sampling for biological and chemical parameters by use of sample return (Breier et al., 2020; Photo credit M. Saito).

When sampling and return are incorporated into expeditions that traverse ocean basins, diagnostic maps across space and time can be created, as demonstrated by the WOCE and GEOTRACES programs, and as one of the field elements planned by the emerging BioGeoSCAPES program (Saito et al., 2024). New generations of robotic sampling systems have been developed to enhance collection abilities such as autonomous underwater vehicles (AUVs) that sample directly (e.g., *Clio*, Jakuba et al., 2018; Breier et al., 2020) or serve as platforms for autonomous samplers (e.g., *Mesobot*; Yoerger et al., 2021; Govindarajan et al., 2023, and the LRAUV-ESP; Truelove et al., 2022). Recently even autonomous surface vehicles (ASVs) with autonomous samplers have been deployed, decoupling oceanographic sampling from oceanographic cruises (Preston et al., 2023). Deployment of currently available underwater sensors for a broad range of important analytes cannot match the spatial coverage and depth of diagnostic capability that high-throughput sampling, return, and analysis can currently accomplish. *Clio*, for example, is a full-depth (6000 m) fully autonomous robot that has four large payload bays allowing collection of large-volume filtered samples and filtered and

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unfiltered waters (Figure 3; Breier et al., 2020; Jakuba et al., 2018). Recent use has demonstrated its robustness in collecting hundreds of station/sample depths and thousands of subsamples per expedition, enabling efficient basin-scale sampling of a multitude of complex biochemical samples (Cohen et al., 2024). *Clio* also provides new sampling capabilities that are not possible with wire-based deployments, including high-resolution vertical sampling at chlorophyll maxima, chemoclines, and nepheloid layers near the seafloor. Similarly, the midwater vehicle *Mesobot* has payload capacity for large-volume samplers, and sampling strategies can take advantage of *Mesobot's* Lagrangian nature to collect time-series data (Govindarajan et al., 2023). Integration of sample return onto diverse robotic, towed, and moored platforms, each of which offers unique strengths and contextual sensor data, will enable a broad range of scientific questions to be addressed.

Medicine Lacks Comprehensive In-Clinic Diagnostics Despite Greater Resources

To offer some perspective on the challenge of 'imminent and inevitable' in situ sensors, we compare progress with the state-of-the-art in biomedicine, where the biomedical financial investment is orders of magnitude greater than environmental research investment. Despite this financial difference, the presence of analytics in the general practitioner's office or over-the-counter at the pharmacy remains constrained to a small number of simple physical measurements (e.g., blood pressure, oxygen saturation, temperature) and specific test kits (e.g. disease antigen kits and glucose monitoring). The use of at-home test kits has proliferated recently due to the COVID-19 pandemic, yet major investment is required to produce each reliable assay. Instead, the biomedical industry continues to rely heavily on blood collection facilities (phlebotomy) and sample shipment to massive centralized laboratories with advanced analytical equipment to achieve efficiency and economies of scale. In other words, the medical industry also relies on the sample collection, return, and laboratory analysis model for many of its critical analytical parameters, despite benefiting from orders of magnitude greater investment.

Improving the strategy for biomedical diagnostics is of course of great commercial interest. Notably, there was the recent spectacular failure of the Theranos company that aimed to develop an inexpensive multi-parameter analysis capability to be deployed within doctors' offices and pharmacies using pinprick-sized samples. Their research and development foci even overlapped with the technologies used in the ocean space such as microfluidics and proteomics techniques described above. Despite enormous investment, the company was

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unable to deliver an in-the-clinic laboratory product, leading to bankruptcy and even fraud convictions. One of the lessons learned from this episode described in the Journal of Clinical Chemistry and Laboratory Medicine was to "be clear what your technology can do today – not what you hope it can do someday" (Fiala and Diamandis 2018). In summary, even the large medical industry still lacks widespread use of in-clinic diagnostics. And this is before adding the additional engineering challenges of operating corrosive, biofouling seawater environments at underwater pressures. Similar lessons can be adapted for the ocean science community: where we continue to pursue our current and future goals for ocean observing without overanticipating or emphasizing particular methods of data collection, e.g., not relying solely on an idealized future full of AUVs equipped with in situ sensors capable of any analyte. Instead, we as a community should broaden our approach to include a range of data collection approaches. Doing so recognizes that we are already currently at the cusp of routinely fielding a range of new robotic observing platforms, equipped with sensors and samplers, capable of leveraging both the best available sensing technology and results derived from the most advanced laboratory capabilities currently available.

Combining Ocean Vital Signs and Diagnostics

There is increasing interest in the study of ocean health and vitality (Halpern, 2020; Fernandez, 2022), with considerable investment in sensor-based applications. When proposing to complement in situ sensors with sample return capabilities, a critique we repeatedly hear is that sample return does not have sufficient resolution relative to the sampling by sensors (T, S, O_2) deployed by floats, vehicles, and moorings to enable statistical interpretation and modeling. We argue that this sensors-are-all-we-need argument is perhaps short-sighted. Can a sensors-only approach to the question of ocean health produce the best information needed for detecting ocean change and informing policymakers?

We can explore this using the human health analogy. When you go to the doctor's office they initially rely on the in-office measurements of vital signs such as temperature, blood pressure, and oxygen saturation - note some similarities to the key CTD sensor parameters here. Would you feel confident that that was sufficient information to understand your health and offer to forgo the use of phlebotomy blood sampling and laboratory analyses? You could request greater replication of those three parameters to improve confidence and allow statistical analysis, such as thousands of temperature or oxygen measurements, even scaling up to continuous monitoring with wearable sensors. Would those sensors be able to identify the

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underlying cause of a patient's illness, such as cancer or a specific infection? Most would agree that low-cost sensors measure *vital signs* that can observe health conditions such as fevers, high blood pressure, and low blood oxygen. Yet they lack the resolving power of complementary laboratory *diagnostics*, like cancer biomarkers, vitamin levels, cholesterol, and hormones to name a few analytes routinely measured from a single blood sample with great economies of scale. In general, in the clinic sensors are used as rapid, low-cost indicators of vital signs of overall health that are complemented by information-rich laboratory diagnostic measurements that can determine the underlying cause.

Similar analytical power is available now for natural environments, often using the same analytical technology. We understand the complementary power of laboratory analyses in human health settings, especially when coupled with advanced sample return capabilities as an integrated analytical strategy. Why would we shun their use in ocean and planetary health, especially if they can be globally deployed with economies of scale? We contend that the notion that sample return has insufficient sampling resolution to be useful or scalable is short-sighted. It depends on the question being asked and the purpose of the replication. This argument also discounts the accumulation of observations across multiple deployments, regions, and time periods that develop into comprehensive datasets capable of supporting statistical analysis of environmental change. In ocean settings, transect spacing of several degrees using nutritional biomarkers has been shown to be a reliable means to detect primary and co-limiting controls on ocean productivity across ocean basins (Saito et al., 2014; Cohen et al., 2024). Higher spatial or temporal sampling could be employed to understand process effects such as the influence of storms or eddies. Often a stated end goal in oceanographic projects is to produce new or improve existing computational models. However, it is worth arguing that, similar to the clinic, laboratory diagnostics of environmental stress do not necessarily require a computational model to be interpreted. Indeed, diagnostics can be used to validate and improve biogeochemical general circulation model performance and forecasting capability.

Conclusion

To be clear, we are not arguing that sensors lack value: they have an important role in environmental research and should continue to be developed. Instead, we are documenting what is intuitive to many chemists, oceanographers, and ecologists: there are fundamental chemical, biochemical, and engineering reasons that make inexpensive, robust in situ analyses difficult to accomplish for many analytes. As we envision oceanographic research and

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monitoring in the coming decades, it is prudent to remind ourselves that a dearth of diverse, robust biological and chemical sensors is due to the reality that the analytical task at hand is orders of magnitude harder than finding a needle in a haystack. While sample return lacks the allure of real-time analysis, it is a proven approach that is efficiently and globally scalable by coupling autonomous field sampling approaches to high-throughput laboratory analytical methods. Together, sample return and laboratory analysis provide information-rich diagnostic data that complement the sensor-based vital signs information.

Author contribution and **Competing interests**

Saito and McIlvin conceived of the perspective. Saito weighed the needles and conducted calculations. All authors discussed the concepts and contributed to the manuscript writing.

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Data Availability

Data used for calculations is available within supplemental materials.

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