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Revisiting five decades of ^{234}Th data: a comprehensive global oceanic compilation.

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Abstract. We present here a global oceanic compilation of ^{234}Th measurements that collects results from researchers and laboratories over a period exceeding 50 years. The origin of the ^{234}Th sampling in the ocean goes back to 1967, when Bhat et al. (1969) initially studied ^{234}Th distribution relative to its parent ^{238}U in the Indian Ocean. However, it was the seminal work of Buesseler et al. (1992) - in which it was proposed that particulate organic carbon (POC) flux could be calculated from ^{234}Th distributions if the ratio of POC to ^{234}Th measured on sinking particles (POC:^{234}Th) at the desired depth was known - that drove the extensive use of the ^{234}Th-^{238}U radioactive pair to evaluate the efficiency with which photosynthetically fixed carbon is exported from surface ocean by means of the biological pump. Since then, a large number of ^{234}Th depth profiles have been collected using a variety of sampling instruments and strategies that have changed the past 50 years. The present compilation is made of a total 223 datasets: 214 from studies published either in articles in referred journals, PhD thesis or repositories, and 9 unpublished datasets. The data were compiled from over 5000 locations spanning all the oceans for total ^{234}Th profiles, dissolved and particulate ^{234}Th concentrations, and POC:^{234}Th ratios (both sediment traps and filtration methods that include two sizes classes; 1-53 µm and < 53 µm). A total of 379 oceanographic expeditions and more than 56000 ^{234}Th and 18000 ^{238}U data points have been gathered in a single open-access, long-term and dynamic repository. This paper introduces the dataset along with informative and descriptive graphics. Appropriate metadata have been included, including geographic location, date, and sample depth, among others. When available, we also include water temperature, salinity, ^{238}U data and particulate organic nitrogen data. Data sources and methods information (including ^{238}U and ^{234}Th) are also detailed along with valuable information for future data analysis such as bloom stage and steady/non-steady state conditions at the sampling moment. The data are archived on PANGAEA repository, with the dataset's DOI doi.pangaea.de/10.1594/PANGAEA.918125 (Ceballos-Romero et al., 2021). This provides a valuable resource to better understand and quantify how the contemporary oceanic carbon uptake functions and how it will change in future.

1 Introduction

For several decades, radioactive tracers have been used to gain a better understanding of different oceanographic processes. In the context of the Biological Carbon Pump (BCP) (Eppley and Peterson, 1979; Volk and Hoffert, 1985), radionuclides such as ^{210}Po and thorium isotopes, are extensively used to study the various physical, chemical or biological processes involved in

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the particle export and flux attenuation in the oceans (Cochran, 2003). Radionuclides are characterized by a unique property: their half-lives, which accounts for the time it takes for one-half of the atoms of a radioactive element to undergo radioactive decay and thus transforming to a different isotope. Half-lives are not affected by temperature, physical or chemical state, or any other influence. As a result, the concentration of radioactive elements varies over time at well-characterized decay and production rates. Observations of radionuclides distributions in the water column, in time and space, provide valuable insights into the presence and rates of ocean processes on spatial scales from local to basin-wide and timescales of days to months and years depending on the radionuclide employed.

The naturally occurring radioisotope ^{234}Th has been commonly used to understand natural aquatic processes in four major areas: vertical transport, particle cycling, horizontal transport, and sediment dynamics (Waples et al., 2006). ^{234}Th is collected by a variety of sampling procedures since its initial used by Bhat et al. (1969). ^{234}Th chemistry dictates that it is adsorbed onto particles surface and effectively scavenged from the dissolved water phase. Hence, when biological activity is high, a ^{234}Th is removed from the surface ocean and transported downward by the sinking particles, thereby generating a deficit relative to its soluble or "conservative" parent ^{238}U. This deficit can be used to calculate the downward flux of ^{234}Th. An excess in ^{234}Th activity – i.e. a daughter concentration higher than the parent one - is attributed to fragmentation processes that result in the conversion of sinking to non-sinking particles, generically termed "remineralization" (Maiti et al., 2010). Due to its short half-life of $T_{1/2}$ = 24.1 days (decay constant: $\lambda = \ln 2/T_{1/2} = 0.02876$ d^{-1}, mean life: τ = 34.8 d) and its particle reactivity in seawater it is suitable to trace processes occurring in the upper ocean on time scales from days to months (Rutgers van der Loeff et al., 2006), or even shorter when high scavenging by particles (Turnewitsch et al., 2008) (see Sect4.2).

^{234}Th has been an indispensable tool in many oceanographic field expeditions. The most widespread application of the ^{234}Th approach is to estimate the gravitational settling of carbon as Particulate Organic Carbon (POC) out of the surface layer, which results in a downward flux of POC (e.g., Bacon et al., 1976; Ceballos-Romero et al., 2016; Kirk Cochran et al., 1993; Rutgers van der Loeff and Geibert, 2008; Stewart et al., 2011; Verdeny et al., 2009). To a lesser extent, this radionuclide is also used to estimate the downward flux of other elements to the deep ocean, such Particulate Inorganic Carbon (PIC) (e.g., Le Moigne et al., (2013a); Wei et al., (2011)), Biogenic Silica (BSi) (e.g., Buesseler et al., (2005); Le Moigne et al., (2013a)), Particulate Organic Nitrogen (PON) (e.g., Buesseler et al., 1992; Charette and Buesseler, 2000; Murray et al., 1996) or trace metals fluxes (e.g., Black et al., 2018; Weinstein and Moran, 2005).

It was in the 90's that an increasing number of studies for ^{234}Th took place. This increase in use is in part due to a variety of new sampling instruments and strategies that have changed over the years. In 2006, a special issue entitled Future Applications of ^{234}Th in Aquatic Ecosystems (FATE, https://www.sciencedirect.com/journal/marine-chemistry/vol/100/issue/3) edited by Edited by Claudia R. Benitez-Nelson, Robert F. Anderson was published in Marine Chemistry with the purpose of thoroughly reviewing the use of ^{234}Th in aquatic systems. Papers included reviews of the applications and future uses of this radiotracer (Benitez-Nelson and Moore, 2006; Waples et al., 2006), discussions on the techniques and methodologies used for ^{234}Th analyses (Rutgers van der Loeff et al., 2006), the impact of POC:^{234}Th ratios and their sampling methodology on POC flux

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estimates (Buesseler et al., 2006), and ^{234}Th sorption and export models in the water column (Savoye et al., 2006), among other topics. As one of the most actively used tracers in oceanography, Waples et al. (2006) already reported 237 papers dealing with ^{234}Th published in refereed journals. However, after 5 decades of extensive use, a unique repository of ^{234}Th measurements has never been compiled and ^{234}Th data remain scattered when not belonging to major sampling programs. Major observational programs like JGOFS (Joint Global Ocean Flux Study, http://usjgofs.whoi.edu/), LTER (Long-Term Ecological Research, https://lternet.edu/), UKOA (UK's Ocean Acidification, https://www.bodc.ac.uk/projects/data_management/uk/ukoa/cruise_programme/), AMT programme (Atlantic Meridional Transect, https://www.bodc.ac.uk/projects/data_management/uk/amt/), CHOICE-C (Carbon Cycling in the China Seas: Budget, Controls and Ocean Acidification, http://coe.xmu.edu.cn/En/ContentShow.aspx?Id=COEE_3_3), GEOTRACES (https://www.geotraces.org/) or, most recently, EXPORTS (EXports Processes in the Ocean from RemoTe Sensing, https://oceanexports.org/projectoffice.html); or seminal field experiments such as VERTIGO (VERtical Transport In the Global Ocean, https://cafethorium.whoi.edu/projects/vertigo/), SoFeX (Southern Ocean Iron Experiment, https://cafethorium.whoi.edu/projects/sofex/), EisenEx (European Iron Enrichment Experiment in the Southern Ocean, https://www.bco-dmo.org/project/2056), or CHINARE (Chinese National Arctic Research Expedition, https://arcticportal.org/ap-library/news/2046-journal-advances-in-polar-science-sees-rapid-development-and-progress-of-chinare-program-on-arctic-science) have never been brought together for ^{234}Th results. Therefore, it is valuable to bring together all existing ^{234}Th concentration data, along with appropriate metadata, in one repository to facilitate further use and analysis.

Previous efforts compiling ^{234}Th-based data have been created to access only ^{234}Th-derived POC fluxes (see Le Moigne et al., 2013b) and, more recently, POC:^{234}Th ratios (see Puigcorbé et al., (2020) and doi.pangaea.de/10.1594/PANGAEA.911424). In contrast, ^{234}we have compiled the complete results of ^{234}Th concentration measurements in sea water and particles at every depth, location and time of sampling. The compilation can be found in doi.pangaea.de/10.1594/PANGAEA.918125. This article is the report of the compilation, a unique dataset to better understand and quantify how the contemporary oceanic carbon uptake functions and how it will change in future.

The goal of this effort is to serve as a basis for an open-access, long-term and dynamic oceanic repository of ^{234}Th measurements and useful metadata to be used in an accessible, easily findable, and inter-operable way that grows from now on from the contribution of other authors involved in ^{234}Th sampling. Moreover, given the great number of metadata and parameters compiled, the compilation offers multiple ways to use ^{234}Th, even opening the possibility of exploring new applications. For that reason, we have chosen not to compile the derived parameters reported by other authors, such as ^{234}Th downward fluxes or ^{234}Th-derived POC fluxes, but rather provide the data necessary for others to make such analyses, open to the criteria, modelling, and interpretation chosen by each researcher.

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2 Data

2.1 Data organization

We have gathered datasets consisting of ^{234}Th measurements in oceanic waters sampled between 1967 and 2018. The compilation includes a total of 56482 data points for ^{234}Th concentrations, distributed as follows: i) 21458 for total, ii) 6591 for dissolved, iii) 13955 for particulate ^{234}Th measurements, and iv) 10851 and v) 3631 for POC:^{234}Th and PON:^{234}Th ratios, respectively. Additionally, ^{238}U (18257 data), POC (7659 data) and PON (3624 data) concentrations are also reported. Temperature and salinity values are also included when possible, making a total of 5959 values complied for temperature and 12722 values for salinity. When fields are missing for a given record, the data are entered as "-999".

The data have been extracted from a total of 219 different studies published in refereed journals between 1969 and 2020, 4 PhD dissertations, 8 datasets accessible in public repositories, and 9 unpublished ones directly provided by authors (listed in *Table 1*) spanning all oceanic regions (*Figure 1a*) and compiled in a total of 223 datasets. The data repositories include: BCO-DMO® ("Biological & Chemical Oceanography Data Management Office", https://www.bco-dmo.org/), DARWIN® ("Data and Sample Research System for Whole Cruise Information in JAMSTEC", http://www.jamstec.go.jp/e/about/informations/notification_2021_maintenance.html), EDI Data Portal® ("Environmental Data initiative", https://portal.edirepository.org/nis/home.jsp), and PANGAEA® (https://www.pangaea.de/). Additionally, data corresponding to published GEOTRACES® cruises can be found in GEOTRACES website (Intermediate Data Product 2017 (Schlitzer et al., 2018)).

The data sets are provided in both a total of 223 excel spreadsheets and a single compilation file, hosted in PANGAEA repository (doi.pangaea.de/10.1594/PANGAEA.918125). Each spreadsheet is univocally identified with a unique integer record ID number (denoted as "Reference_USE") and consists of two tables: i) the "metadata" and ii) the "data".

2.1.1. "Metadata" sheet

The "metadata" table is a list of the data origin in its broadest sense at a glance. It contains information structured in 6 categories: 1) "REFERENCE_USE", 2) "INFO", 3) "DATA", 4) "METHODS", 5) "ADDITIONAL_DATA", 6) "DATA_SOURCE". The full list of metadata included in each dataset and a brief description of the table fields can be found in the Supplementary Material (SM) (see *Table S1)*.

There are data from 379 cruises, covering 5134 locations spanning all oceanic regions (*Figure 1a*). Some stations were part of cruise transects whereas others were part of small-scale surveys or reoccupation of the same location at different seasons and years. In all cases, sampling region and period – including bloom stage at the sampling moment when indicated by the authors - are given as metadata as part of the "INFO" section. A summary of the sampling period, the maximum and minimum latitude and longitude of the region surveyed, and the maximum depth sampled for ^{234}Th is provided. Blooms stages are distinguished as "non-bloom", "pre-bloom", "bloom" and "post-bloom" conditions, respectively. Additionally, information such as the

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project name, when sampling took place within a major observational program, cruise name, leg details, research vessel, and chief scientist are also indicated. A total of 17 fields are reported in this section (see *Table S1*).

The "DATA" section provides information of the dataset contents at glance with YES/NO indicators to the basic data of ^{238}U, ^{234}Th (total, dissolved and particulate phases) and POC(PON):^{234}Th ratios, the fraction size(s) details, and the number of total stations and samples reported. A total of 13 parameters are detailed in this section.

The "METHODS" section is intended to provide useful interpretive information regarding how the sampling and/or measurements were accomplished at a glance. It provides basic information about i) ^{238}U determination: whether it was directly measured, or salinity derived, in which case the salinity relationship employed is specified; ii) total ^{234}Th sampling and radiochemical purification methods, iii) dissolved, and iv) particulate ^{234}Th phases; and v) the modelling approach followed when ^{234}Th data were used to estimate POC fluxes (i.e., the assumption of steady (SS) or non-steady state (NSS) conditions). A final space for comments of any kind is included in this section.

We also report the existence of additional data of interest in the "ADDITIONAL_DATA" section with YES/NO indicators for the cases of i) ^{234}Th underway sampling, ii) sediment traps deployments, iii) ^{234}Th was paired with ^{210}Pb-^{210}Po disequilibrium sampling, and iv) CHN (Carbon-Hydrogen-Nitrogen). Note that this section is merely intended as informative of the sampling methods used complementarily to the ^{234}Th technique. Both ^{234}Th underway data and POC:^{234}Th and PON:^{234}Th ratios from sediment traps are reported when available. However, neither 210Pb-210Po concentrations nor CHN data are compiled in this dataset. The availability of these data is indicated as reported by authors.

Finally, several details regarding the data source are included in the 8 fields specified in the "DATA_SOURCE" section, including a YES/NO indicator for the publication date. The data owners are always clearly indicated by the first author of the publication or dataset. In the case of data published in research articles, the journal and the publishing year are indicated, while in the case of unpublished data obtained from personal communication (as is the case for 9 datasets), "np" (stating for non-published) is given in the journal information and no data are provided as publication year. Where data had been assigned a Digital Object Identifier (DOI), this is included in DOI/others. When no DOI is available, the data URL source –either database or publication links, is included instead. Other URL sources or personal communication from data source is keyed by the text variable "data_resource" in the "metadata" table, when available.

Most of the measurements were obtained from publications. In these cases, if the data were transcribed from tables, the table number is also given as "data localization". If data were only available graphically, a computer program to digitize the data from plots was used (WebDigitizer, https://automeris.io/WebPlotDigitizer/) and the figure number is also given. In the rare cases in which data were not accessible through any of these procedures, the authors were directly contacted for data is indicated in the localization.

Finally, a space for further links or information of interest is provided under the text variable "other DOI/resources". In the few cases that the same data were reported in another publication (e.g., Murray et al., (1996), Dunne et al., (1997) and Murray

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et al., (2005) reported the same data from the U.S. JGOFS program during 1992 in the Equatorial Pacific), it was indicated under this text variable.

2.1.2. "Data" sheet

The "data" table is the dataset core and contains all the data detailed above (more details in *Table S2*).

Each data point is accompanied by cruise and station IDs, location – latitude, longitude, depth, and additionally bottom depth when available – and sampling date – including month-date-year and Day of Year (DOY) formats –. All ^{234}Th (total, dissolved and particulate activity) concentrations were converted to dpm L^{-1} (density1027 kg m^{-3}) if not already reported in these units. POC:^{234}Th and PON:^{234}Th ratios are given in µm dpm L^{-1} and include samplings with sediment traps and filtration methods, in which we included i) bottles- Go-Flo and Niskin types- ii) filtration systems, iii) SPLITT (split flow-thin cell fractionation), and iv) (large or small volume) in-situ pumps for either the entire particulate fraction or two sizes classes (preferably 1-53 µm and < 53 µm) when available. These size-classes were chosen largely based on results of Bishop et al., (1977), Clegg and Whitfield (1990), and others, who assumed that the >53 µm size-class is responsible for most of the mass flux into traps. Other cut offs of 51 µm or 70 µm and other size classes are found and are noted when different than 53 µm. The particles sampling method - categorized as "method 1" (for filtration methods) and "method 2" (for sediment traps) -and size fractions – categorized as "small" or "large" - are specified as part of the data. Total, particulate and sediment traps ^{234}Th sampling depths are separately indicated. When reported, water temperature, salinity, and ^{238}U measurements are included. Moreover, if accessible, POC and PON concentrations (in µm L^{-1}) are included.

In all cases we assume that the originating authors and editors have undertaken steps necessary to control data quality. Measurement uncertainties in the data points as provided by the data measurer (e.g., "uncert_total_^{234}Th"). Please refer to original source for whether data uncertainty includes only the one signa counting error, or other factors, such as uncertainty on volumes, detector efficiency, background, etc.

2.2 Data formats and availability

The data are archived on PANGAEA repository, with the dataset's DOI https://doi.pangaea.de/10.1594/PANGAEA.918125 (Ceballos-Romero et al., 2021). The data table is available for download either as a unique merged file containing all data sets and metadata or as individual excels files.

Moreover, the template followed to compile the dataset (including the "metadata" and "data" tables) along with instructions to fill in this template are made available in PANGAEA for any author who either wants to review, complement a dataset included in this compilation or contribute to its extension with a new dataset.

Finally, a *suggestions box* has been made available at PANGAEA for authors to submit suggestions or request to amend the datasets compiled.

3 Scope and introduction to the dataset

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In this article, we aim at providing a broad overview of the character of the datasets to be used with different purposes in future studies. We therefore provide several graphics to indicate the scope and nature of the data compiled. These include maps of sampling locations (*Figure 1a*) and time histories of analyses per year for ^{234}Th measurements (*Figure 2*) among others. Additionally, a summary table with the most remarkable metadata is also provided in doi.pangaea.de/10.1594/PANGAEA.918125. Note that the compilation here presented does not overlap the previous compilation by Le Moigne et al. (2013), where exclusively ^{234}Th-derived POC fluxes were gathered together and mostly for a fixed depth of 100 m or 150 m only.

Here, we give an overview of the compilation and some important aspect for its use. A historical review of the 50 years of the ^{234}Th studies is given in Sect 4, while some of the perspectives and applications we suggest for the datasets are briefly commented on in Sect 5.

3.1. General overview

The sampling locations shown in *Figure 1a* are dominated by cruise tracks mostly in the Northern Hemisphere (NH) (*Figure S1* in the SM), although there are also a few locations in the Southern Hemisphere (SH) with significant repeated sampling, especially in the Southern Ocean (*Figure 1b)*. Over a total of 379 cruises compiled, 294 took place in the NH (78%), only 53 in the SH (14%) and a total of 32 (8%) crossed the Equator and sampled locations in both hemispheres.

Many of these expeditions were carried out in the framework of lager research programs. For those cases, this information has been included as metadata in the compilation. During these 50 years of study, we have identified a total of 14 major ocean programs that have provided invaluable knowledge to the ^{234}Th approach (see *Table 2*). A total of 68 (30%) of the datasets were collected during surveys as part of one of these 13 programs, as reported by the authors. Additionally, for cruises belonging to specific projects or experiments, this information has also been compiled.

Sampling locations for ^{234}Th also include a number of long-term, high-frequency observations at fixed locations in the open ocean, referred to as (long-term) time-series stations, TSS- from now on. These stations offer a crucial observational strategy for capturing the dynamic temporal changes in ocean conditions and biogeochemical processes at the seasonal and decadal scale. Moreover, it allows investigating short-term episodic events usually unaccounted for by oceanographic cruises. A great number of time-series stations that are either operational, registered (for future operation), inactive or closed spread across the globe (source: https://www.ocean-ops.org). We have acknowledged the importance of sampling at these TSS by identifying matches with ^{234}Th sampling locations (see *Figure 1c)*. Coincidences between locations with ^{234}Th data available and the position of a time-series station have been reported in the metadata section with the label “time-series” in the “number of stations” field. Note that this should not be mistaken with repeated sampling of a location during a cruise, which has also been acknowledged with the label “re-occupied” in the same field. A total of 98 datasets (44%) reported samplings on at last one TSS location while only 33 (15%) reoccupied the sampling site. The most frequent TSS sampling sites found in the compilation include the *Bermuda Atlantic Time Series Study* (BATS: 31.50N, 64.10W, http://bats.bios.edu/), the *Hawaii Ocean Time series*

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(HOT: 22.45N, 158W, https://hahana.soest.hawaii.edu/hot/hot_jgofs.html) at station ALOHA, the *Dynamics of Atmospheric Fluxes in the MEDiterranean* sea (DYFAMED: 43.42N, 7.87E, http://www.obs-vlfr.fr/cd_rom_dmtt/sodyf_main.htm) – all of them part of JGOFS -, and some other relevant ones such as the *South East Asia Time Series* (SEATS: 18.00N, 166.00E, https://www.odb.ntu.edu.tw/seatsen/#:~:text=The%20SEATS%20time%2Dseries%20station,Arctic%20Sea%20is%20the%20largest), also from JGOFS, the *Palmer Antarctica* and *California Current Ecosystem Long-Term Ecological Research Project* (PAL-LTER (https://pal.lternet.edu/) and CCE-LTER (https://cce.lternet.edu/) respectively), or the *Porcupine Abyssal Plain* (PAP, 49.00N, 16.50W, https://projects.noc.ac.uk/pap/) site sustained observatory among others (*Figure 1c)*. More details of the history of these locations repeatedly sampled for ^{234}Th will be given in forthcoming sections.

Over time, the interdisciplinary scope of the studies and the number of techniques and parameters simultaneously assessed have gradually evolved. Field surveys have changed not only in terms of number but also in terms of strategy (e.g., duration, spatial resolution and repeat occupation of the same site). This has influenced not only the number of data points measured – with a sustained increasing trend - but also the type of ^{234}Th data reported (i.e., total, dissolved and particulate phases, and POC(PON) to ^{234}Th ratios). Similarly, it has driven changes in the number of ^{234}Th studies published. We have identified 3 key indicators of these inflection points. In the 90's the number of ^{234}Th studies dramatically increased after the publication of the first empirical routine method to estimate downward POC fluxes in the ocean by Buesseler et al. (1992). Another tipping point took place in the 2000's likely due to significant improvements in the ^{234}Th methodology derived from the introduction of the small volume technique (Benitez-Nelson et al., 2001b). Finally, a later shift took place with the launching of GEOTRACES in 2010, which changed the way ocean in currently explored and expanded applications to trace metals and isotopes. More details of these milestones and the consequences they brought to the ^{234}Th technique will be discussed in Sect 4.

The overall temporal evolution of ^{234}Th measurements is depicted in *Figure 2*. *Figure 2a* shows the annual number of field expeditions including ^{234}Th sampling per sampling year between the initial measurements in 1967 to 2018, separated by oceans. *Figure 2b* shows the number of ^{234}Th-derived export data presented per year, including the 9 data sets previously unpublished and made accessible through this compilation. And *Figure 2c* shows the histogram of ^{234}Th data points sampled since 1967 separated by data type. Additionally, the annual oceanic distribution of ^{234}Th data points is shown in *Figure S2*.

Despite the fact that the first ^{234}Th study took place in the Indian Ocean in 1967, this ocean along with the waters from the Arctic Ocean, visited by the first time in 1994, is the least surveyed one during this 50-year compilation (*Figure 2a*). The Atlantic and Pacific oceans dominate the sampling locations, followed by the Southern Ocean, to a much lower extent. Field expedition to Pacific Ocean started in 1970, in 1977 to the Atlantic and in 1987 to the Southern Ocean. Other locations include several rivers, lakes, and bays (see *Figure 1a*).

During the 60's-70's, expeditions including ^{234}Th measurements were scarce, with the total number of annual samplings never exceeding 5. It was in the 90's when the number of ^{234}Th samplings started to dramatically increased, with an average number

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of annual expeditions of 15. In the subsequent years, a great number of cruises took place, with an average of 10 per year during the last two decades. However, the annual number of dedicated cruises is not significant of the number of ^{234}Th points reported by year. In the past, the number of cruises including ^{234}Th measurements greatly increased over time, and so did the number of data points, while more recently, a larger number of data (*Figure S2*) are reported from a reduced number of cruises (see *Figure 2a)*.

There are several remarkable peaks in the annual record of ^{234}Th data points which are related to dedicated carbon export programs and experiment that would be detailed in next sections. A delay of about 3-4 years relative to sampling is commonly observed for the publication of these ^{234}Th data and derived results, started in 1969, as depicted in *Figure 2b*, where the number of ^{234}Th-derived export data per year is shown, including the 9 data sets previously unpublished and made accessible through the compilation presented here. Namely, as shown in *Figure S2*, peaks in the number of data points stand out in: i) 1992, when the JGOFS equatorial Pacific process study took place, published by several groups in 1995-1997 (Bacon et al., 1996; Buesseler et al., 1995; Dunne et al., 1997; Murray et al., 1996) and extended in 2001 with the publication of POC:^{234}Th ratios from sediment traps results by Hernes et al., (2001); ii) 1997, when the JGOFS Southern Ocean study and the Arctic expedition ARK XIII/2 were carried out, published by Buesseler et al., (2001b) and Rutgers van der Loeff et al., (2002b) respectively; iii) 2004, when several intense expeditions took place, with VERTIGO voyages as the most productive ones in terms on data contribution, published by Buesseler et al., (2009) and Lamborg et al., (2008b); the Shelf-Basin Interactions (SBI, https://arctic.cbl.umces.edu/sbi/web-content/) Phase II field program, published in 2007 by several authors (Lalande et al., 2007; Lepore et al., 2007 and Lepore and Moran 2007); the EDDIES project, synthesized by Buesseler et al., (2008a); and the CROZEX project available in Morris et al., (2007), iv) 2010, where a total of 17 cruises took place, with the Southern Ocean cruises (Owens, 2013) as the most intense ones #in term of number of ^{234}Th samples per day and the several GEOTRACES section cruises: 2 cruises to the South Atlantic Ocean as part of UK GEOTRACES published by (Le Moigne et al., 2014), 2 U.S. GEOTRACES and 1 Dutch GEOTRACES published by Owens et al., (2015) and 2 cruises in the Atlantic Ocean from 64°N to the equator published by (Puigcorbé et al., 2017a); and finally v) 2018, when 2 GEOTRACES cruises (data compiled in GEOTRACES data base and PANGAEA) and the first field expedition of the EXPORTS program took place (published by Buesseler et al., (2020)).

3.2. Total, particulate and dissolved ^{234}Th

Changes in the type of ^{234}Th data measured during filed expeditions are depicted in *Figure 2c*. While within the first years of the ^{234}Th technique, authors focused on the total, dissolved and particulate phases mainly for the study of the chemical scavenging of ^{234}Th and the partitioning of Th species between phases (see e.g., Bacon and Anderson, 1982; McKee et al., 1986; Murray et al., 1989; Nozaki et al., 1981), in the 90's the broader potential of ^{234}Th and its application as a tracer for particle dynamics and export fluxes became evident, which drove the inclusion of sampling strategies for the determination of POC(PON) to ^{234}Th ratios. Given the reduced annual number of data points in the 70-80`s, in comparison to subsequent years, it is difficult to perceive the contribution of each data type to the total number of data points within the first years of study. A

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detailed review of the evolution of ^{234}Th data available thought years in the framework of the mentioned milestones is given in Sect 4.

In summary, total ^{234}Th is almost always reported by all authors along years by either direct measurement or determined as the sum of the dissolved and particulate phases. It is worth mentioning that the distinction between dissolved and particulate is operational. Dissolved and particulate specie have traditionally been discriminated by filter sizes, typically of 0.2-l µm pore size (Moran and Buesseler, 1993), with the submicron colloidal matter - nanometer to submicrometer size range ~0.001-l µm (Stumm, 1977) - included in the dissolved phase. Note that colloidal size ranges have not been included in the compilation but indicated in the metadata section when available. A general view of the ^{234}Th particulate data compiled is provided in *Figure S3*, which shows the locations with ^{234}Th concentrations sampled on either small (generally from 0.7-1 to 53-70 µm) large (>53 or >70 µm) or both size fractions. An overall 75% of the studies compiled reported particulate ^{234}Th concentrations, more than half of them (56%) reported data in 2 size fractions.

In a reduced number of studies, ^{234}Th concentrations were determined as a single vertically integrated sampled, normally between 0 and 100 m depth (i.e., Buesseler et al., 1998, 1994, 1995; Charette et al., 1999; Charette and Buesseler, 2000; Cochran et al., 2000; Evangeliou et al., 2011; Hall et al., 2000; Ma et al., 2005; Maiti et al., 2008; Schmidt et al., 2002). This sampling approach was deliberately done to reduce sample numbers, yet maximize location sampled, at a time when at sea analyses was more difficult. This has also been indicated both in the metadata and data sections in each individual spreadsheet with the code number "-555" in the depth column (see *Table S1* and *Table S2)*.

A total of 66% of the studies reported POC:^{234}Th ratios measured with some of the filtration methods (described in detail in Sect 2.1.2), while only ~16% reported PON:^{234}Th ratios. In ~40% of the studies compiled, POC(N):^{234}Th ratios collected with sediment traps were reported from a great variety of trap designs: Indented Rotating Sphere (IRS), surface-tethered, free-floating, bottom-moored, automated, cylindrical, VERTEX-style, U-type, CLAP-type, RESPIRE type, Free-drifting Lagrangian Sediment Traps (LST), High Frequency Flux (HFF) standard drifting particle interceptor traps (PITS) and, more recently, neutrally buoyant sediment traps (NBSTs) and Particle Export measurement using a LAGRAngian (PELAGRA) trap. An overview of POC:^{234}Th data compiled is shown in *Figure S4*.

Note that a specific, more detailed, compilation of global POC:^{234}Th ratios in the ocean has been recently published by Puigcorbé et al (2020) (archived in the data repository PANGAEA® under doi.org/10.1594/PANGAEA.911424) with the purpose of elucidating the spatial, temporal and depth variations of this crucial parameter. The authors present a database of 9318 measurements sampled on 3 size fractions (~> 0.7 µm, ~ 1–50 µm, ~> 50 µm) collected with in situ pumps and bottles and bulk particles collected in sediment traps from the surface down to > 5500 m. Our compilation includes the studies gathered in Puigcorbé et al (2020) and expands the dataset with a total of 10851 POC:^{234}Th ratios.

3.3. ^{238}U measurements

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Finally, in order to allow the assessment of POC fluxes, ^{238}U concentrations has been also included. This is generally reported by authors either by direct measurement or, most commonly, derived from salinity data by applying one of the ^{238}U-salinity relationships available. From the studies compiled, we have identified a total of 10 salinity-^{238}U relationships published in journal articles, chronologically ordered as follows: i) Ku et al., (1977); ii) Broecker and Peng (1982), iii) Coale and Bruland, (1985), iv) Chen et al., (1986), v) Andersson et al., (1995), vi) Delanghe et al., (2002), vii) Rutgers van der Loeff et al., (2006), viii) Pates and Muir (2007), ix) Owens et al., (2011), x) Martin et al., (2013), along with some other relationships also frequently used such as 2.4 dpm/kg at salinity 35, 0.06813 x salinity ‰, or 0.07097 x salinity ‰. The most extended ^{238}U-salinity relationships are the ones given by Ku et al. (1977) (A_U = 0.07081 × salinity), Chen et al. (1986) (A_U = 0.0686 × salinity) and, more recently, by Owens et al. (2011) (A_U (±0.047) = 0.0786 x salinity – 0.315). Furthermore, metadata of the model assumptions – categorised as Steady (SS) and non-steady state (NSS) (more details in Sect 4.2.)– and the stage of the bloom have also been included (as "bloom", "pre-bloom", "post-bloom" and "no bloom"), when available for the sake of a better interpretation of the data for the accurate POC flux assessment, as recommended by Ceballos-Romero et al. (2016), (2018).

4. Discussion: ^{234}Th timeline

The compilation covers a temporal range of 5 decades. The temporal distribution of oceanic ^{234}Th measurements begins in 1969 to measure its distribution in the hydrologic cycle (Bhat et al., 1969) and extends up to these days as an indispensable tool in a oceanographic field expedition. We consider it was not the passing of years but rather the publication of seminal key studies that delineates the progression of ^{234}Th studies. For this reason, we have divided the ^{234}Th technique time history in four well distinguished eras.

4.1. Era 1: "The Old Man and the Sea"

The first era was initiated by the study of Bhat et al. (1969), in which a total of 6 profiles of total ^{234}Th:^{238}U ratios in depths up to 250 m were collected on-board the U.S.C. and G.S.S. Oceanographer during 1967 at several sampling sites in the Indian Ocean to study scavenging processes. Polyvinylchloride (PVC) tube samplers of 30 L capacity designed by Shale J. Niskin (Niskin, 1962) were used. This pioneering study by Bhat was expanded by many other investigators (e.g., Kaufman et al., (1981); Knauss et al., (1978); Lee et al., (1991); Matsumoto (1975); Tanaka et al., (1983); Tsunogai et al., (1986)). Bhat et al. (1969) assumed that essentially all ^{234}Th in seawater was in particulate form, whereas Matsumoto (1975) assumed particulate ^{234}Th to be an insignificant part of the total ^{234}Th. The analysis of particulate ^{234}Th was incorporated by Krishnaswami et al., (1976) during a survey in the Pacific Ocean. Several authors combined total and particulate ^{234}Th concentrations (e.g., Dominik et al., (1989); Minagawa and Tsunogai (1980); Santschi et al., (1979)). Finally, the sampling of the ^{234}Th dissolved phase was initiated by McKee et al. (1984), who collected samples in coastal environments near a major sediment source (Yangtze River). But it was the influential study of Coale and Bruland (1985) the first one that presented several profiles of ^{234}Th in both dissolved and particulate form for the sake of elucidating the partitioning of ^{234}Th between these phases, which allowed the

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authors to demonstrate that this radionuclide is an ideal particle reactive tracer for studying the scavenging of thorium from surface waters.

In fact, a series of papers by Coale and Bruland in the mid-80's (Bruland and Coale, 1986; Coale and Bruland, 1985, 1987) were key for establishing the baseline for future ^{234}Th studies that would use ^{234}Th as a proxy for POC fluxes in the next era. The authors discovered that the ^{238}U-^{234}Th disequilibrium is a direct tracer of the rates of sinking particles from the upper ocean, realizing the relevance to the downward ^{234}Th flux. Previously, on a one-year time-series study, Tanaka et al. (1983) found large variations in the total ^{234}Th activities, with the minimum in ^{234}Th inventory coinciding with the early spring bloom, which they proposed was related to biological activity in the surface waters. Following these studies, Eppley et al. (1989) proposed that, if ^{234}Th is scavenged by biogenic particles, ^{234}Th could be used as a tracer of export production.

The "Coale and Bruland papers" were followed by many other investigators (e.g., Bacon and Rutgers van der Loeff (1989); Huh and Beasley (1987); Kershaw and Young (1988); Murray et al. (1989); Rutgers van der Loeff and Berger (1991); Schmidt et al. (1990); Wei and Murray (1991)). Additionally, the initiation of global ocean chemistry, hydrographic, and tracer survey efforts that took place within this era, especially GEOSECS (the GEOchemical Ocean SECtions Study, http://iridl.ldeo.columbia.edu/SOURCES/.GEOSECS/), from 1972 to 1978) and the first years of JGOFS (from1987 to 2003) contributed to further increased ^{234}Th research and pointed towards the second era of the ^{234}Th technique.

A series of milestones have been identified as the most remarkable of this era, which summaries as follows:

- 1969: initial measurement of total ^{234}Th and introduction of the co-precipitation of ^{234}Th with $Fe(OH)_3$ (Bhat et al., 1969).
- 1972: beginning of GEOSECS program, which would prologue until 1978 and transformed the field of chemical oceanography as it existed at that time by exploiting new technologies available then (see synthesis of results by Broecker and Peng (1982).
- 1984:
  - initial analysis of particulate and dissolved ^{234}Th in river (McKee et al., 1984);
  - introduction of the MnO2-impregnated filter cartridges technique (Mann et al., 1984), used with in-situ pumps.
- 1985: initial analysis of particulate and dissolved ^{234}Th in open ocean and link between biological processes and ^{234}Th deficits clearly demonstrated (Bruland and Coale, 1986; Coale and Bruland, 1985, 1987).
- 1987:
  - Joint Global Ocean Flux Study (i.e., beginning of the JGOFS era, which would last until 2003);
  - 1 long-term time-series established: *DYFAMED: Dynamics of Atmospheric Fluxes in the MEDiterranean sea* (43.42N, 7.87E) in the frame of JGOFS France program.

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- 1988: 2 long-term time-series projects established: *BATS: The Bermuda Atlantic Time-series Station* in the Atlantic Ocean (31.50N, 64.10W), and *HOT: Hawaii Ocean Time-series* in the Pacific Ocean at Station ALOHA: A Long-term Oligotrophic Habitat Assessment (22.45N, 158W) in the frame of U.S. JGOFS program.
- 1989: ^{234}Th proposed to trace export production (Eppley et al., 1989).

There are at present two distinct radioanalytical methods for the ^{234}Th extraction and purification from water samples that were introduced during this era: i) the co-precipitation of ^{234}Th with $Fe(OH)_3$, introduced by Bhat et al., (1969), and ii) the scavenging of this nuclide onto MnO_2 cartridges, introduced by Mann et al., (1984). A thorough review of these techniques can be found in Rutgers van der Loeff et al., (2006). Briefly, for the $Fe(OH)_3$ technique, 20-30L sea-water samples are treated and beta-counted. The addition of Fe carrier forms a precipitate that serves for removing Th (and other radionuclides) from solutions, so ion exchange purification procedures are required (at sea or quickly after return to shore) to separate ^{234}Th from its parent and other potential beta emitters. For the MnO_2 technique, seawater is sequentially pumped through filters and two MnO2 impregnated cartridges (MnA and MnB) connected in series to scavenge dissolved Th isotopes and gamma counted. This technique was often used with large volume samples (10^2-10^4L), needed primarily for ^{228}Th, ^{230}Th and ^{232}Th analyses, which required large amounts of ship time given the use of in-situ pumps for filtration, limiting the spatial coverage of the ^{234}Th profiles. MnO_2 and Mn cartridges do no adsorb appreciable ^{238}U, which is another advantage as ingrowth after sampling from ^{238}U can be neglected. The large samples represented by a Mn cartridge also allowed for direct gamma counting, thus eliminating the need for laborious radiochemical purification.

A wide variety of methods were used to sample the different ^{234}Th phases within era 1. For the total phase, PVC tube and Van Dorn samplers (i.e., horizontal water bottle) were the prevalent equipment (Bhat et al., (1969); Matsumoto, (1975); McKee et al., (1984) or Minagawa and Tsunogai (1980) among others), although a few studies used pumping systems (Lee et al., 1991; Tsunogai et al., 1986), and bottles (Bacon and Rutgers van der Loeff, 1989). Volumes between 12 and 800L were sampled. In the majority of the studies measuring dissolved and particulate ^{234}Th phases, the total ^{234}Th concertation was estimated as the sum of them (see e.g., Coale and Bruland (1985); Murray et al., (1989). Regarding particulate ^{234}Th analyses, most of the studies used filtration systems on volumes between 30 and 700L, a few studies used bottles (with Go-Flo model more typical than Niskin one), and one study introduced the use of pumps (Bacon and Rutgers van der Loeff, 1989), with the predominant particulate size class cut-off was 0.45 µm. Note that the distinction between the dissolved and particulate phases is generally operational, with the term "dissolved" usually comprising all the phases passing through a 0.4 µm Nuclepore filter (McKee et al., 1986).

A total of 25 published works in refereed journals comprise the ^{234}Th studies of this era, compiled in a total of 24 datasets. Sampling was characterized by cruises mostly in the Pacific Ocean (see *Figure 2a* and *Figure 3a*), with a reduced number of locations sampled (maximum of 29 locations and averaged value of 6 locations per cruise) and a reduced number of data points per study (rarely over 100, averaged value of 60 samples per cruise). A total of 36 cruises were compiled from this era, with a

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total of 2177 data points. Most surveys took place in the NH (29) between April and September (*Figure 4a*). Only 1 expedition surveyed the SH, while a total of 6 cruises crossed the Equator and sampled in both hemispheres.

Only 2 studies reported samplings that were part of major programs, which included the GEOSECS program (Krishnaswami et al., 1976) and the DYFAMED program (see *Table 2* for a summary). Another study took place within the framework of the Joint Chinese-American Field Program (JCAFP) (McKee et al., 1984). A total of 4 studies reported sampling time-series stations. Except from Tanaka et al., (1983) that applied a non-steady state model to estimate the residence time of ^{234}Th, all the studies that provided information in this regard, assumed steady-state conditions during sampling, which is not surprising since stations were not usually reoccupied during cruises for the collection of time-series data.

During this era, it became very common to separate ^{234}Th into of several phases. This is specifically significant for the dissolved phase, whose sampling would have less interest over time. A reason for this is that initial ^{234}Th studies were focused on analyzing the parameters influencing the partitioning of a species between dissolved and particulate phases as a way to understand the mechanisms and rates for the scavenging of particle-reactive species. One way to quantify the scavenging of particle-reactive species is the use of distribution coefficients (K_d), which measure the partitioning of a species between dissolved and particulate phases (McKee et al., 1986). As the knowledge of this processes increased, novel applications of ^{234}Th were developed, driving changes in the ^{234}Th data type collected during field expeditions (see next sections).

The majority of the studies reported ^{238}U concentrations along with at least one ^{234}Th phase concentration, but none of them measured POC:^{234}Th (*Figure 5a*), as the importance of this parameter was not evinced until era 2 (see Sect 4.2). ^{238}U was either measured (a total of 6) or derived from salinity (a total of 10) using the relationship from Ku et al., (1977). More than half of the studies (60%) modelled ^{234}Th data, with all of them (a total of 14 studies) but one assuming SS conditions. It is also worth mentioning that very few studies reported simultaneous sampling of complementary non-thorium measurements, such as sediment traps (a total of 6 studies) or 210Pb-210Po disequilibrium (5 studies) or CHN data (3 studies).

4.2. Era 2: "The voyage of the beagle"

A transcendental boost to the number of ^{234}Th measurements occurred within what we are calling the "JGOFS era" (1989-2003). In 1992, a new approach was used to estimate POC fluxes from ^{234}Th distributions, starting with the U.S JGOFS North Atlantic Bloom Experiment (NABE). During this study, the seminal work of Buesseler et al. (1992) found a clear relationship between the onset of the spring bloom, the subsequent drawdown of nutrients and CO_2, the net removal of ^{234}Th and the flux of POC. Because of the conservative nature of ^{238}U in the ocean, any measurable deficit of ^{234}Th relative to it can be assumed to imply a significant removal by scavenging and particle sinking flux over a period of days to weeks before sampling, as the response time of ^{234}Th is dictated by ^{234}Th decay constant (λ) and removal rate by particles (k): 1/(λ+k) (Turnewitsch et al., 2008). Accordingly, Buesseler et al. (1992) postulated the empirical determination of POC export fluxes from ^{234}Th-^{238}U oceanic disequilibrium following

$$POC_{flux}(z) = POC{:}234\mathrm{Th}(z) \cdot 234Th_{flux}(z) \quad . \tag{1}$$

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Where *POC:* ^{234}Th is the ratio of POC to ^{234}Th measured on sinking particles at the desired depth *z* (in µmol dpm^{-1}), and $^{234}Th_{flux}$ is the ^{234}Th downward flux measured at the same depth.

Eq. (1) is the base of the so-called ^{234}Th method and the POC flux obtained is referred to as ^{234}Th-derived POC flux. Note that the concept for this empirical method was introduced in a conference abstract in one much earlier study in the North Pacific (Tsunogai et al., 1976). Buesseler et al. (1992) proposed that the sinking flux of any element – such as carbon, phosphorus or nitrogen - could be derived from ^{234}Th flux if the ratio of this element to ^{234}Th on sinking particles is known. *POC:element* ratios are directly determined from its in-situ measurement and vary with both depth and particle size dependent (Buesseler et al., 2006). ^{234}Th flux can be calculated from the disequilibrium between the ^{238}U-^{234}Th resulted from particle sinking by evaluating the change in the corresponding total ^{234}Th activity in time and the contributions due to horizontal and vertical advection and diffusion processes of the dissolved ^{234}Th. The simplest solution to estimate ^{234}Th flux is SS conditions and ignoring adjective and diffusive transport. These assumptions are the most commonly used (Le Moigne et al., 2013b) as only single ^{234}Th profile is generally measured. Alternatively, a NSS model, can be applied when activity gradients in time can be assessed, when repeated sampling at the study area, ideally over the course of 2 to 4 weeks, are available for the collection of time-series data (Buesseler et al., 1992; Savoye et al., 2006).

This 1992 publication motivated an increase in oceanic ^{234}Th measurements in the 1990's and for that reason, it was chosen as the beginning of era 2 in the ^{234}Th timeline, which we consider lasting until 2001. It was characterized by the following key milestones:

- 1992: introduction of the empirical method for the POC flux estimate from ^{234}Th concentrations proposed by Buesseler et al., 1992.
- 1992-1993: studies of the role of colloidal material (i.e., ~0.001 > colloids < l µm (Stumm, 1977)) in ^{234}Th scavenging (Baskaran et al., 1992; Moran and Buesseler, 1993).
- 1995:
  - introduction of the use of the ^{210}Pb-^{210}Po pair in a similar manner to the ^{238}U-^{234}Th pair by Shimmield et al., (1995);
  - development of a regional 3-D ^{234}Th flux model to estimate the vertical and horizontal gradients of ^{234}Th (Buesseler et al., 1995).
- 1996: beginning of the expansion of the ^{234}Th-approach to other elemental fluxes (e.g., Particulate Inorganic Carbon-PIC) (Bacon et al., 1996).
- 1999:
  - introduction of the 20-L MnO2 co-precipitation technique for ^{234}Th by Rugters van der Loeff and Moore (1999). This technique allowed analyzing the particulate and dissolved activities of ^{234}Th in a single aliquot, thus enabling the calculation of total ^{234}Th activity (particulate plus dissolved) as well as its residence time in the two phases.

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Moreover, it enabled on board beta counting for ^{234}Th with a portable beta counter, leading to obtain higher temporal and spatial resolution;

- 1 long-term time-series established: *SEATS: South East Asia Time-Series Station* (18N, 116E) in the frame of JGOFS France program.

In the earlier experiments of this era, bottle POC data were generally compared to ^{234}Th-derived from individual cartridge filters or other particle collectors, such as in Buesseler et al., (1992). Several options of large volume in situ pumps that allowed for measurement of POC and ^{234}Th on the same filter emerged during this era: i) large volume filtration using Challenger Oceanic's submersible pumps was introduced by Shimmield et al., (1995), ii) MULVFS (multiple-unit large-volume filtration system) in situ pumping system by Charette et al., (1999), with large volumes (2500-3500L) passed sequentially through a 1 µm cartridge pre-filter we initiated, and iii) large volume (400L) samples collected using in situ battery-operated pump deployed on the CTD/Rosette frame and filtered (Charette and Buesseler, 2000). Ratios were normally determined using small pored sized filters, either >0.4-0.5 µm (Cochran et al., 1995; Guo et al., 1997; Huh and Prahl, 1995; Santschi et al., 1999), or >1 µm (e.g., Benitez-Nelson et al., 2000; Gustafsson et al., 1997; Rutgers van der Loeff et al., 1997) typically made of glass or quartz fiber. Only 7 studies sampled 2 size fractions, adding POC:^{234}Th ratios on large particles, determined by filtering through 53 µm (Buesseler et al., 1998; Moran et al., 1997) or 70 µm Nitex screen (Charette and Buesseler, 2000; Cochran et al., 2000). Additionally, to a lesser extent, POC:^{234}Th ratios in bulk particles were collected in sediment traps, including modern designs such as neutrally buoyant sediment trap (NBST) and surface-tethered particle interceptor trap (PIT).

A total 42 datasets comprise this era, extracted from a total of 47 publications. Sampling was focused on the Atlantic Ocean (see *Figure 2a and Figure 3b*). Surveys in the Pacific Ocean were limited while increased sampling in the Southern Ocean, and field expeditions to the Arctic were conducted. Both the number of expeditions and the locations sampled increased. Despite its short duration (less than a decade), a total of 70 cruises were reported within this era, indicative of the dedicated programs and experiments that marked this era, resulting in 9872 data points. Similar to the first era, cruises mainly took place in the NH (52 cruises), although expeditions to the SH increased (7 in total) and 11 cruises collected sampled in both hemispheres. Samplings mainly took place within the first semester of the year (*Figure 4b*).

As previously mentioned, this period of the ^{234}Th history partially overlapped with the golden years of the JGOFS era, and therefore, many of the field expeditions took place in the frame of this international program. In the case of U.S. JGOFS, this included ^{234}Th measurements in the BATS and HOT long-term time-series (see section 4.1) and during several process studies in well-defined boxes at strategic oceanic locations: 1) NABE (North Atlantic Bloom Experiment, http://usjgofs.whoi.edu/research/nabe.html) that was one of the first major activities of JGOFS and took place in 1989 along longitude 20°W, 2) EqPac (Equatorial Pacific, http://usjgofs.whoi.edu/research/eqpac.html) process study that was conducted along 140°W during year 1992 and included a total of 4 cruises with the objective of determine the fluxes of carbon and related elements, and the processes controlling these fluxes between the Equatorial Pacific euphotic zone and the atmosphere and deep ocean, 3) Arabian Sea (http://usjgofs.whoi.edu/research/arabian.html), beginning in October 1994 and ending in January 1996,

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4) AESOPS (Antarctic Environment and Southern Ocean Process Study, http://usjgofs.whoi.edu/research/aesops.html), which carried out field work between August 1996 and April 1998. Additionally, studies also reported data from other JGOFS expeditions from the: 5) Indian program, which completed three major sampling expeditions for ^{234}Th to the eastern and central Arabian Sea in April-May, 1994, February-March, 1995 and July-August, 1995, 6) Canadian program in the northeast Pacific Ocean and had two phases, from 1992 -1994 and from 1995-1997, although ^{234}Th was only sampled during the second phase, and 7) France DYFAMED program (http://www.obs-vlfr.fr/cd_rom_dmtt/sodyf_main.htm), which was established in the late 80's (see section 4.1). Finally, filed work also included 8) the Southern Ocean Iron RElease Experiment (SOIREE, https://www.bco-dmo.org/project/2051), which was the first in situ iron fertilization experiment performed in the polar waters of the Southern Ocean. It took place in February 1999 south of the Polar Front in the Australasian-Pacific sector of the Southern Ocean. Data from this iron enrichment experiment was compiled along with others in a common open-access database during Iron Synthesis program (FeSynth, https://www.bco-dmo.org/program/2017) started in 2007.

In addition to JGOFS, cruises from another major initiatives that included ^{234}Th sampling was the Ocean Margins Program (OMEX, http://po.msrc.sunysb.edu/omp/), a large field-based study, with extensive physical, chemical, biological and geological measurements, made on northwest European shelf break that ran in two phases, from 1993-1996 and from 1997-2000. A summary of these activities is provided in *Table 2*.

The number of data points measured per study significantly increased within this era, especially for the dissolved and particulate phases, whose measurements increase more than a 5-fold relative to those from era 1 (see *Figure 5b*). Additionally, measurements to determine POC and PON to ^{234}Th ratios became routine, therefore allowing the estimate of POC fluxes (see *Figure 3b*). However, only half of the studies reported ^{238}U concentrations along with ^{234}Th data, in their majority using a variety of ^{238}U-salinity relationships, with Chen et al., (1986) as the prevalent one. An overall 79% of the studies (38 out of 47 that comprises this era) modelled ^{234}Th data. Time-series data were collected more often during cruises than during the first era, likely with the purpose of following the NSS approach (Buesseler et al., 1992, 1995) - although the majority of the studies (70%) that modelled ^{234}Th data assumed SS conditions during sampling, a total of 11 studies applied the NSS model, with 7 of them combining it with the SS model (e.g. Buesseler et al., (1994); Cochran et al., (2000); Kersten et al., (1998); Wei and Murray, (1992)). In terms of additional data, 1 study introduced underway ^{234}Th sampling (Hall et al., 2000), a total of 10 studies reported sampling with sediment traps and for 210Pb-210Po disequilibrium, and 32 studies measured CHN data.

4.3. Era 3: "In the heart of the sea"

The final boost to the widespread and increasing use of ^{234}Th as a particle flux tracer was motivated by the works of Buesseler et al., (2001) and Benitez-Nelson et al., (2001), which dramatically increased the number of ^{234}Th measurements and marked the third era in the ^{234}Th timeline with the introduction of the small-volume (2-4L) technique. This procedure modified the 20 L-method developed by Rugters van der Loeff and Moore (1999) in era 2 and uses the lowest sample volumes of all known

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^{234}Th methods to date. It not only allowed immediate on-board beta-counting of ^{234}Th activity, but also enhanced both spatial and temporal resolution of particle export and, for that reason, we have chosen it as a shifting milestone.

The revolution that this novel technique brought with it was substantial. The small-volume technique is essential to capture the particle dynamics and export flux variations on scales that could be better related to local biogeochemical conditions. The main advantage is the convenience of handling small volumes and more rapid processing times. Multiple sampling casts are often required for a 20-L sample profiles whereas a 4-L technique usually allows simultaneous sampling of ^{234}Th with other parameters on a single cast (e.g., nutrients, phytoplankton biomass, etc.). As a result, this method can be easily applied at sea using samples obtained through CTD rosette water samplers that are available on most research vessels. Hence, this method not only allowed immediate on-board beta-counting of ^{234}Th activity, but also enhanced both spatial and temporal resolution of particle export along with other biogeochemical parameters. The further improvements of this technique carried out some years later by Pike et al. (2005) and Cai et al., (2006), marked the major milestones of this era, which lasted until 2010 and whose timeline summaries as follows:

- 2001: introduction of the small-volume technique by Buesseler et al., (2001) and Benitez-Nelson et al., (2001).
- 2002: 1 long-term multidisciplinary and moored observatory established: *PAP site: Porcupine Abyssal Plain* (49N, 13.5E), which is the closest abyssal environment to Northwest Europe and has produced a time-series dataset for over 20 years coordinated by the *National Oceanography Centre*. Note that full-depth mooring was established in 2012, with autonomous sensors to measure ocean temperature, salinity, chlorophyll fluorescence, carbon dioxide and nitrate.
- 2005: introduction of the use of yield tracers - double spike technique - for the sake of maintaining the reproducibility (Pike et al., 2005). The authors proposed an acidification of the samples followed by an addition of ^{230}Th as a yield monitor to trace co-precipitation of ^{234}Th by MnO_2 and a second spike (^{229}Th) to correct for losses of ^{230}Th during ion-exchange chemistry. Both ^{230}Th and ^{229}Th are measured using ICP-MS. This double spike technique is standardly used and is recommended by GEOTRACES protocol (Maiti et al., 2012). The advantage of this modified approach is a precise knowledge of ^{234}Th recovery, leading to an enhancement in data quality of ^{234}Th.
- 2006: reduction of the filtration time and introduction of the alpha spectrometric measurement of ^{230}Th recovery by means of using a combination of water bath heating and a reduction in reagent quantities (Cai et al., 2006). This also modified the typical ion-exchange chemistry to allow for the alpha spectrometric measurement of ^{230}Th recovery.

An additional improvement of the method has been recently carried out by Clevenger et al., (2021), which introduces a revised protocol that decreases sample volumes to 2 L, shortens wait times between steps, and simplifies the chemical recovery process, expanding the ability to more rapidly and safely apply the ^{234}Th method.

Moreover, a technological innovation was introduced during era 3 with the development of 2 in-situ pumping systems for high volume filtration for ^{234}Th contraptions and POC:^{234}Th and ratios in sinking particles widely used from this era on: 1) McLane large-volume water transfer system (WTS-LV) pumps by McLane Laboratories (Falmouth, MA), and 2) SAPS (Stand-Alone

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Pump System) by Challenger Oceanic (Surrey, U.K.). Moreover, one study reported the use a novel split flow-thin cell fractionation (SPLITT) (see Gustafsson et al., 2006).

This has been the golden age of the ^{234}Th so far in terms of number of samples, with 76 datasets extracted from 87 studies (82 publications in referred journals, 1 PhD thesis, and 4 repositories) and 4 surveys carried out during those years gathered in the compilation presented here. A total of 159 cruises and 24213 data points were reported in a period of 9 years (see *Figure 3c* and *Figure 5c*). Sampling in the context of major programs continued (i.e., HOT, BATS, OMEX and DYFAMED), and some other new studies initiated, such as the CArbon Retention In A Colored Ocean (CARIACO, http://www.imars.usf.edu/cariaco) time series program operative between 1995 and 2017, or the High Latitude Time Series Observatory project (HiLaTS) established in 2001 by the Japan Marine Science and Technology Center (JAMSTEC, http://www.jamstec.go.jp/e/) at 3 sites (K1; 51N 47E, K2; 47N 60E, and K3; 39N 169E). Additionally, novel JGOFS initiatives took place: 1) JGOFS-France MEDATLANTE (http://www.obs-vlfr.fr/cd_rom_dmtt/other_main.htm) in the Gibraltar Strait and the N.E. Atlantic Ocean, 2) JGOF-France ANTArctic RESearch program (ANTARES, http://www.obs-vlfr.fr/cd_rom_dmtt/an_main.htm), 3) JGOFS-France DYNAPROC (http://www.obs-vlfr.fr/cd_rom_dmtt/other_main.htm) cruise, and the 4) U.S. JGOFS-Japan North Pacific Process Study carried out in at KNOT station (44N 155E, https://www.nodc.noaa.gov/archive/arc0013/0001873/1.1/data/1-data/general/res_outline/KNOT.html) (see *Table 2* for more details). And a cruise was reported as part of the pilot GEOTRACES program, which will be fully discussed in next section as it was chosen as the trigger of the fourth era of ^{234}Th (see Sect 4.4). Finally, a great number of projects and experiments belong to this era (see *Table S3* for a chronological summary).

Samplings spanned the entire ocean and intensified in all regions, except the Indian Ocean (see *Figure 2a and Figure 3c*). Expeditions to the SH increased, with 18 reported, but yet were far below those in the NH, where a total of 134 cruises were undertaken (see *Figure 4c)*. A total of 11 surveys collected sampled in both hemispheres. Stations included the long-term observatories previously sampled for ^{234}Th along with novel ones, such as the PAP site (Turnewitsch and Springer, 2001). Fieldwork took place throughout the entire year, with a remarkable number of cruises in March. Both the overall number of data points and the data points measured per cruise increased due to the large number of field expeditions carried out during this era and the spread use of the small-volume technique. An average of 250 ^{234}Th data points was reported per study. As for the ^{234}Th sample types in this era, measurements increased significantly for all phases (2-fold for the dissolved and particulate while and 4-fold for the total), and for POC:^{234}Th ratios, and remain constant for PON:^{234}Th (see *Figure 5c)*.

A great increase in the number of data points also was detected for ^{238}U concentrations, with an increase of 4-fold in the data points. Once again, salinity-derived was the most extended approach, with almost all the studies using Chen et al., (1986).

A total of 75 studies out of the 87 that comprise this era (i.e., 86%) modelled ^{234}Th data: 50 of them applied the SS model, 2 applied the NSS one and 23 studies applied both the SS and NSS approaches (e.g., Amiel and Cochran, 2008; Buesseler et al., 2001b; Gustafsson et al., 2004; Kim and Church, 2001; Pinghe et al., 2001).

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In terms of sampling strategy, 1 study carried out ^{234}Th underway sampling (Schmidt et al., 2002a). 31 studies reported the used of sediment traps, although only 13 of them used them to measure POC:^{234}Th ratios (Amiel et al., 2002; Aono et al., 2005; Coppola et al., 2002; Hernes et al., 2001; Santschi et al., 2003). Half of the studies reported measurements CHN data, while a very reduced number, less than a 10% of the studies, analyzed 210Po (e.g., Rutgers van der Loeff, 2007; Somayajulu et al., 2002).

4.4. Era 4: "20,000 legs under the sea"

This era is identified with the beginning of internationally dedicated large-scale collaborative projects characterized by long field surveys with very high spatial resolution and many different parameters measured simultaneously. We have marked the beginning of this era in 2010, with the launching of GEOTRACES program (https://www.geotraces.org/) - an international study of the marine biogeochemical cycles of trace elements and their isotopes - which changed the way to explore the oceans by combining ocean sections, process studies, data synthesis, and modeling.

The history of the GEOTRACES dates back to 2000, when the initial idea started with group discussions at international meetings inspired by both the successes and the limitations of GEOSEC and JGOFS. After some years in the planning and enabling phase, the GEOTRACES Science Plan was published in 2006. During 2007-2009 the first GEOTRACES cruises took place, including those that were part of the International Polar Year (IPY, https://www.geotraces.org/geotraces-in-the-international-polar-year/) devoted to detecting and understanding a suite of trace elements and isotopes in the Arctic and Antarctic marine environments, and the intercalibration cruises (to the Atlantic and Pacific oceans in 2008 and 2009 respectively). A GEOTRACES transect in the Drake Passage was also carried out two years later in the framework of IPY (Expedition Zero and Drake). Finally, the GEOTRACES program formally launched its seagoing effort in January 2010 and was fully announced to the scientific community at Ocean Sciences Meeting that year in Portland, OR. Numerous curses have taken place since then, with many of them sampling ^{234}Th as reported in the Intermediate Data Product, the first one released in 2014 (Mawji et al., 2015) and the second one in 2017 (Schlitzer et al., 2018).

This era includes the development of new technologies to accelerate the collection and analysis of samples, the intercalibration of those technologies to ensure internal consistency among the participating labs, the development of a data management system to facilitate access to the results to the entire oceanographic community, and a broad collaborative effort to model, synthesize, and interpret the results. So rather than one specific event like in the previous eras, we identify as the milestone of this era, the new philosophy that the GEOTRACES program brought. It drove a shift from mostly deriving POC fluxes only to include trace metal fluxes. Furthermore, a standards and intercalibration initiative was implemented to establish procedures and protocols for sampling at sea that will ensure that samples are collected, handled, and stored without contamination or other sources of bias (see Cutter et al. (2010) and Maiti et al., (2012) for ^{234}Th), which are the most noteworthy milestones on this era. We specially acknowledge the efforts by Cutter et al. (2010) to ensure that ^{234}Th results produced by different groups

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were comparable and internally consistent, using deep waters or stored samples as standards where ^{234}Th and ^{238}U and known to be in secular equilibrium.

A total of 81 datasets were compiled from this era, including 9 previously unpublished ones, extracted from a total of 72 studies (65 publications in referred journals, 3 PhD thesis and 4 data repositories). A total of 114 cruises and 38476 data points were compiled, mostly distributed between the Atlantic and Pacific oceans and, to a lesser extent, the Southern Ocean (see *Figure 2a*). Once again, the NH dominated the surveys (79 cruises in total and an additional 8 sampling also below the Equator), particularly numerous in early spring and fall (see *Figure S1*). Nonetheless, this era reports the highest amount of cruises to the SH, with a total of 27.

More than half of the expeditions of this era took place in the framework of a major ocean program (see *Table 2*) and some minor projects, such as the Arctic Ocean 2001 (AO-01) expedition, the 2nd 3rd Chinese National Arctic Research Expedition (CHINARE-2 and 3), the Indo–German iron fertilization experiment LOHAFEX (https://epic.awi.de/id/eprint/21440/), the second KErguelen Ocean and Plateau compared Study expedition (KEOPS2), or the Fate of the northwestern Mediterranean open sea spring bloom (FAMOSO, http://digital.csic.es/handle/10261/94572) project. Furthermore, a 46% of the datasets included ^{234}Th data from an established time-series location (see *Figure 1c*).

An interesting characteristic of this period is that a relatively smaller number of cruises produced a greater amount of data in comparison to previous eras. The most relevant peaks are found in years 2013 and 2015, which coincides with the publication of several GEOTRACES transects (see Sect 3). Dissolved ^{234}Th measurements reduced drastically (4-fold), while the rest of measurement types increased to some extent, most notably for total ^{234}Th and ^{238}U (see *Figure 5d*, note the change in the scale of this plot along the eras). Once again, ^{238}U derived from salinity was the most commonly used approach with almost all the studies using the U-salinity relationship of Chen et al., (1986) or (Owens et al., 2011).

In terms of modeling ^{234}Th data, a total of 64 out of the 72 studies (91%) modelled ^{234}Th concentrations and specified whether a SS and NSS model was used. From them, 48 (75%) studies applied the SS approach, just 1 (2%) study applied the NSS one, and a total 15 (23%) studies compared both approaches (see Buesseler et al., 2020; Kim et al., 2011; Lemaitre et al., 2018; Martin et al., 2011; Planchon et al., 2013; Roca-Martí et al., 2017; Rutgers van der Loeff et al., 2011; Zhou et al., 2012 among others). The number of studies carrying out ^{234}Th underway sampling increased to 5 (e.g., Black et al., 2018; Estapa et al., 2015; Martin et al., 2013). More than 41% of the studies reported POC:^{234}Th ratios collected with sediment traps (Haskell et al., 2013; Kawakami et al., 2010; Luo et al., 2014; Maiti et al., 2016). Almost all the studies of the studies reported measurements CHN data (88%), while a very reduced number (~10%) analyzed 210Po (such as Alkalay et al., 2020 or Wei et al., 2011) and only 3 studies simultaneous measured and discussed ^{234}Th-derived results from ^{234}Th, 210Po and compared then to fluxes from sediment traps (Ceballos-Romero et al., 2016; Le Moigne et al., 2013a; and Stewart et al., 2010).

4.5. "Song for the blue ocean": the beginning of era 5?

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Significant milestones within recent years are marked by the launching of an unprecedented number of large-scale collaborative projects including ^{234}Th measurements: e.g. 1) COMICS: Controls over Ocean Mesopelagic Interior Carbon Storage (UK, 2017-2021, https://www.comics.ac.uk/), a research project that aims to quantify the flow of carbon in the ocean's twilight zone (part of the ocean between 100 and 1000 m below the sea surface) in order to more accurately model global climate change; 2) EXPORTS: EXport Processes in the Ocean from Remote Sensing project (US, 2017-2022, https://oceanexports.org/), a large-scale NASA-led field campaign that will provide critical information for quantifying the export and fate of upper ocean net primary production (NPP) using satellite observations and state of the art ocean technologies; 3) CUSTARD: Carbon Uptake and Seasonal Traits in Antarctic Remineralisation Depth project (UK, 2018-2022, https://roses.ac.uk/custard/), an initiative which will examine how seasonal changes in food availability for phytoplankton at a key junction of the global ocean circulation influences how long carbon is trapped in the ocean rather than escape to the atmosphere as carbon dioxide; 4) OTZ: Ocean Twilight Zone (US, 2018-2024, https://twilightzone.whoi.edu/), a project devoted to the study of this region of the ocean, with particular empathizes on development of new technologies for scientific exploration of the biomass and biodiversity, animal lives and behavior, food webs interactions, and flow of carbon through the twilight zone, and 5) SOLACE: Southern Ocean Large Area Carbon Export (Australia, December 2020-January 2021, https://solace2020.net/), a 6-weeks voyage aimed at developing an approach to quantify the changing effectiveness of CO_2 sequestration by the BCP using remote-sensing of the ocean surface by satellites and it's interior by autonomous vehicles (specially BIO-ARGO profiling floats). Additionally, there are other planned initiatives that include ^{234}Th in their methodologies such as the APERO program: Assessing marine biogenic matter Production, Export and Remineralisation: from the surface to the dark Ocean (France, https://www.polemermediterranee.com/Activites-Projets/Ressources-biologiques-marines/APERO) or PICCOLO: Processes Influencing Carbon Cycling: Observations of the Lower limb of the Antarctic Overturning (UK, https://roses.ac.uk/piccolo/). The Joint Exploration of the Twilight Zone Ocean Network (JETZON, https://www.jetzon.org/) was created in 2020 to act as an international coordinating umbrella to serve as a focal point for Twilight Zone studies and it includes ^{234}Th measurements as one of the key methodologies.

Note that so far, only data from the first expeditions of the EXPORTS project are available in the compilation (see peak in *Figure 2b* and *Figure 2d*). The benefits to be derived from the success of these endeavors and the inclusion of the remaining data in the compilation could trigger the beginning of a new era for the ^{234}Th technique (i.e., era 5).

5. Significant gaps in the global dataset

During the last few decades, considerable progress has been made towards unraveling the behavior of the Biological Carbon Pump (BCP) and understanding of the factors influencing the carbon dynamics and the ocean carbon cycle, e.g., primary production, aggregation, ballasting, and the activities of zooplankton and bacteria (see reviews by e.g., Buesseler and Boyd (2009); De La Rocha and Passow (2007); Sanders et al. (2014); Turner (2002)). Strategies have included time-series of process studies in key ocean regions, global surveys of carbon parameters, long-term time-series sites and models and databases built from field observations.

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Capturing the spatiotemporal variability of ^{234}Th concentrations could play a key role on our precise quantification of carbon uptake, storage rates, and subsequent ecosystem impacts. However, there are limitations for ship-based observing strategies. For example, large areas of the ocean started to be sampled only recently for ^{234}Th and there are still several geographical gasps in the oceans. In terms of surveys geographical distribution, the SH remains clearly undersampled. Of the total of 5134 locations compiled, 3351 belong to the NH (which accounts for a total of 65% of the sampling locations) and only 1749 to the SH (35%), with the 34 remaining ones corresponding to the Equator (less than1%). This is the result of the majority of expeditions taking place in the NH, 78% individually and 8% as part of surveys in both hemispheres. *Figure 1a* highlights an especially important gap in the South Pacific region and some additional notable gaps in the dataset in the Benguela system (already included in COMICS project), the Mauritanian upwelling or the southern Indian Ocean.

Moreover, changes in POC flux with depth for carbon budgets and BCP efficiency assessments derived from in-situ measurements are spatially and temporally limited. Single measurements of POC fluxes are routinely extrapolated to larger spatial scales and to longer timescales for regional and global estimates. This use of the data neglects temporal issues and might led to uncertainties in global algorithms describing carbon export, which might have important implications in the carbon budget estimates. ^{234}Th data along with biogeochemical and physical properties records available from time-series stations in multi-disciplinary observatories could be used to assess the regional and temporal evolution of the BCP. *Figure 1c* shows numerous established time-series stations that have never been sampled for ^{234}Th.

In terms of current gaps for application of ^{234}Th for the assessment of POC fluxes, a fundamental issue is whether we are correctly addressing the temporal conditions at the sampling moment when interpreting ^{234}Th deficits (Ceballos-Romero et al., 2018). The accuracy of these POC flux evaluations relies on the correct calculation of the ^{234}Th flux, which is not straightforward given the physical and temporal processes involved in the ^{234}Th activity balance in a given parcel of water. The change in the total ^{234}Th activity in time and the contributions due to horizontal and vertical advection and diffusion processes of the dissolved ^{234}Th must be evaluated for the correct calculation of the ^{234}Th flux (Buesseler et al., 1992). To a first approximation, these contributions due to advection and diffusion processes are neglected (e.g., Ceballos-Romero et al. (2016); Stewart et al. (2007)); or quantified and found negligible (e.g., Morris et al. (2007)). Accounting for the potential temporal variation of ^{234}Th activities is challenging since many cruises do not always allows repeated measurements at the same site. In those cases, a SS approach – i.e., neglecting changes in the ^{234}Th concentrations over time - is inevitably followed to calculate ^{234}Th fluxes. Only when there is a repeated sampling of the study area after a time gap (several days to weeks) changes in the ^{234}Th activity can be determined accurate enough - i.e., uncertainties associated to the measurement are small enough. In this case, it is possible to estimate the temporal change of radionuclide concentration by including a linear correction to the SS solution, known as the NSS approach (Buesseler et al., 2005; Savoye 2006).

Furthermore, due to the inherent persistence of radionuclide deficits within the water column according to their half-lives, radionuclide-derived POC flux estimates are ascribed to previous export events. This means, for example that, depending on the moment of sampling, ^{234}Th concentrations might not represent the reduction in export at the end of a bloom because the

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deficit relative to ^{238}U still persists. Thus, the SS approach could result in under- (early bloom) or over-estimate (post bloom) of the export fluxes (Ceballos-Romero et al., 2018).

The reliability of the SS/NSS approach applied to ^{234}Th deficits to estimate the POC flux depends on the sampling time in relation to the bloom dynamics (Ceballos-Romero et al., 2018). A large proportion of the ocean exhibits seasonal variability in the export within the half-life of ^{234}Th (Henson et al., 2015). Therefore, a thorough evaluation of the information provided by the ^{234}Th deficits in terms of the spatial and temporal scales accounted for is required. The portion of the bloom period accounted by ^{234}Th sampling must be therefore carefully assessed, for example by means of a comparison to seasonal Primary Production or Chlorophyll data, in order to correctly interpret the export results (Ceballos-Romero et al., 2016). This is especially important in those cases where, due to data availability, a SS approach is inevitably followed to calculate ^{234}Th fluxes.

A total 83% of the studies modelled ^{234}Th data and therefore, indicated the assumptions to interpret ^{234}Th deficits. Most of these studies (72%) followed the SS approach, while 24% of them combined it with the NSS one, and only 4% applied only the NSS model. However, overall, only a 36% of the studies compiled provided information regarding bloom conditions and bloom stage during sampling, which prevents us from assessing the reliability of the assumption of SS and NSS conditions to calculate ^{234}Th fluxes. The available information on the bloom timing, and/or peak flux date, and/or duration detailed by station would be required for a quantitate evaluation of the overall accuracy of current ^{234}Th-SS-derived POC flux estimates (Ceballos-Romero et al., 2016, 2018).

Moreover, in terms of temporal distribution, research cruises are frequently conducted between January and October, although data are skewed to early spring and summer (see peak in April in *Figure S1b*). This is especially important for the SH, in the Southern Ocean, where tough wintertime conditions complicate shipboard operations and research cruises are therefore frequently skewed to calmer spring and summer months. This means that sampling in Southern Ocean mostly occur during bloom conditions, which affects the reliability of the SS approach to quantify carbon export from ^{234}Th data (Ceballos-Romero et al., 2018). Furthermore, results in the Southern Ocean are biased, when compared to the results in other areas, as they correspond mainly to POC fluxes in bloom conditions, whereas in the rest of the oceans sampled, pre and post bloom conditions are also included in the global analyses.

Finally, as mentioned before, the increasing interdisciplinary character of field expeditions allows the simultaneous use of multiple sampling methods and techniques whose combined interpretation provide us with better tools for the seasonal study of ocean processes and marine particle dynamics. In the case of ^{234}Th, its potential increases when combined with methods that account for export episodes over different timeframes of a bloom period, such as ^{210}Po-^{210}Pb, or sediment traps (Ceballos-Romero et al., 2016). For that reason, we included information regarding the availability of these other techniques when combined with ^{234}Th sampling in the metadata section. Note that we have only consulted publications, and cruise reports when accessible, to gather information for these metadata so information as to the existence of these data might be missing. We

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therefore recommend using them cautiously when stated "no (available)" but fully trust it when stated "yes". To report an update to the metadata or an error in the data compiled, we encourage authors to contact us.

6. Towards a better understanding of oceanic carbon uptake: data use perspectives

The efficiency of the BCP at transporting carbon to the deep ocean and regulating atmospheric CO_2 levels is affected even by small perturbations to oceanic ecosystems such as seawater chemistry or nutrients distribution (Kwon et al., 2009). Ongoing changes in temperature, oxygen concentration and stratification of the water column derived from anthropogenic warming are predicted to alter export and transfer efficiency by different means. Specifically, a reduction in carbon sequestration, and therefore an increase in oceanic emissions of CO_2 is expected (Kwon et al., 2009; Le Quéré et al., 2009). More observational data and mechanistic models to describe the particle flux and its attenuation are required in order to unravel and properly quantify the current role of the BCP and provide a baseline for future estimates of oceanic CO_2 uptake. ^{234}Th-^{238}U disequilibrium combined with ^{210}Po-^{210}Pb could be used to analyze simultaneously parameters such us export efficiency, transfer efficiency (Buesseler et al., 2020b). Together with particle sinking velocities disequilibrium derived (e.g.Villa-Alfageme et al., 2021) . A multidisciplinary approach that draws knowledge and methodologies from the fields of physics, mathematics, chemistry, biology, oceanography, and marine sciences is necessary. The compilation presented here could be used to provide revised and more robust estimates of the ocean's biological carbon pump by re-evaluating ^{234}Th-derived POC fluxes regionally, by studying the temporal evolution of the BCP strength through time-series data, by studying carbon export at shallow depths, and by globally analyzing the carbon export patterns in relation to the particle sinking velocity or synthesizing modelled particle remineralization.

Data availability

Our dataset is archived in the data repository PANGAEA® (http://www.pangaea.de), under the following DOI: https://doi.pangaea.de/10.1594/PANGAEA.918125 (Ceballos-Romero et al., 2021)

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of the EXPORTS program and WHOI as part of the Ocean Twilight Zone project. M.V.A. and K.O.B. are also part of an IAEA Coordinated Research Project “Behaviour and effects of natural and anthropogenic radionuclides in the marine environment and their use as tracers for oceanographic studies”. The data used are listed in the references, figures, and supporting information.

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Tables

Table 1. Summary of the studies included in the compilation in chronological order of publication. Three types are distinguished as data source type: publications in refereed journals (J), publication in repository (R), PhD thesis (T), and personal communication (P) for unpublished datasets released in this compilation. Note that in some cases, published data have been reported in different publications, for which all the publications are indicated. Basic information of sampling details (i.e., region and period) is also included. Additionally, availability of seven different categories of ^{234}Th data are indicated with an "X" as follows: 1) total ^{234}Th, 2) dissolved ^{234}Th, 3) particulate ^{234}Th, 4) POC:^{234}Th ratios from filtration methods, 5) PON:^{234}Th ratios from filtration methods, 6) POC:^{234}Th ratios from sediment traps, and 7) ^{234}Th underway sampling.

Data source		Sampling details		Data available						
Type	Reference/ Data repository	Sampling region	Sampling period	1	2	3	4	5	6	7
J	Bhat et al., 1969	Indian Ocean	01/01/1967 - 31/12/1967	X						
J	Matsumoto, 1975	Pacific Ocean	01/01/1970 - 31/12/1969	X						
J	Krishnaswami et al., 1976	Pacific Ocean	03/08/1973 - 09/11/1973			X				
J	Knauss et al., 1978	Pacific Ocean	01/01/1973 - 31/12/1973	X						
J	Santschi et al., 1979 & Santschi et al., 1980	Narragnasett Bay	16/01/1978 - 21/02/1979	X		X				
J	Minagawa and Tsunogai, 1980	Pacific Ocean	13/04/1975 - 09/09/1974	X		X				
J	Kaufman et al., 1981	Atlantic Ocean	29/04/1977 - 08/05/1977	X						
J	Tanaka et al., 1983	Pacific Ocean	25/04/1979 - 26/02/1980	X						
J	McKee et al., 1984	Yangtze River China	01/11/1981 - 30/12/1981	X	X	X				
J	Coale and Bruland, 1985	Pacific Ocean	14/11/1978 - 01/09/1980	X	X	X				
J	Bruland and Coale, 1986	Pacific Ocean	01/09/1980 - 09/09/1980		X	X				
J	McKee et al., 1986	Amazon River mouth	08/06/1983 - 10/06/1983		X					
J	Tsunogai et al., 1986	Pacific Ocean	19/05/1981 - 15/03/1982	X						
J	Coale and Bruland, 1987	Pacific Ocean	01/10/1981 - 31/08/1983		X	X				

https://doi.org/10.5194/essd-2021-259
Preprint. Discussion started: 1 October 2021
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J	Huh and Beasley, 1987	Pacific Ocean	01/10/1985 - 30/10/1985		X	X			
J	Kershaw and Young, 1988	Atlantic Ocean	01/11/1985 - 30/11/1985		X	X			
J	Bacon and Rutgers van der Loeff, 1989	Pacific Ocean	03/07/1979 - 17/08/1980	X	X	X			
J	Dominik et al., 1989	Lake Geneva	21/04/1986 - 29/10/1986	X		X			
J	Moran and Moore, 1989	Atlantic Ocean	06/09/1988 - 11/09/1988	X		X			
J	Murray et al., 1989	Pacific Ocean	06/06/1986 - 23/06/1986		X	X			
J	Schmidt et al., 1990	Mediterranean Sea	08/04/1986 - 06/04/1988		X	X			
J	Lee et al., 1991	Pacific Ocean	24/06/1989 - 08/10/1989	X					
J	Rutgers van der Loeff and Berger, 1991	Southern Ocean	07/11/1987 - 09/12/1987	X	X	X			
J	Wei and Murray, 1991	Atlantic Ocean	01/06/1988 - 30/06/1980		X	X			
J	Baskaran et al., 1992	Atlantic Ocean	01/10/1989 - 30/06/1991	X	X	X			
J	Buesseler et al., 1992 & Cochran et al., 1993	Atlantic Ocean	19/04/1989 - 05/06/1989	X	X	X	X	X	X
J	Moran and Buesseler, 1992	Atlantic Ocean	14/05/1991 - 14/05/1991	X	X	X			
J	Sarin et al., 1992	Indian Ocean	01/03/1991 - 30/04/1991	X	X				
J	Schmidt et al., 1992	Mediterranean Sea	17/03/1987 - 14/05/1987		X	X			
J	Wei and Murray, 1992	Dabob Bay	01/01/1987 - 31/12/1987	X	X	X			
J	Baskaran and Santschi, 1993	Galveston coast	01/05/1990 - 31/12/1990	X		X			
J	Moran and Buesseler, 1993	Atlantic Ocean	25/06/1991 - 28/07/1992	X	X	X			
J	Buesseler et al., 1994	Atlantic Ocean	24/04/1992 - 08/05/1992	X			X	X	X
J	Kuptsov, 1994	Arctic Ocean	08/01/1993 - 31/10/1993				X	X	
J	Sarin et al., 1994	Indian Ocean	01/12/1988 - 31/12/1988		X				
J	Sarin et al., 1994a	Indian Ocean	15/03/1991 - 24/02/1992	X					
J	Thunell et al., 1994	Pacific Ocean	07/01/1988 - 02/07/1988			X			
J	Buesseler et al., 1995	Pacific Ocean	01/03/1992 - 02/12/1992		X	X	X		X
J	Cochran et al., 1995	Arctic Ocean	19/07/1992 - 11/08/1993		X	X			

https://doi.org/10.5194/essd-2021-259
Preprint. Discussion started: 1 October 2021
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J	Huh and Prahl, 1995	Pacific Ocean	07/08/1993 - 09/08/1993	X	X	X			
J	Niven et al., 1995	Atlantic Ocean	17/03/1993 - 17/05/1993	X	X	X			
J	Shimmield et al., 1995	Southern Ocean	07/12/1992 - 07/12/1992		X	X			
J	Bacon et al., 1996	Pacific Ocean	22/03/1992 - 21/10/1992	X	X	X			
J	Baskaran et al., 1996	Atlantic Ocean	16/03/1992 - 25/06/1992	X	X	X			
J	Murray et al., 1996 & Dunne et al., 1997 & Murray et al., 2005	Pacific Ocean	07/02/1992 - 13/09/1992	X	X	X	X	X	X
J	Sarin et al., 1996	Indian Ocean	12/04/1994 - 12/08/1995	X	X				
J	Guo et al., 1997 & Santschi et al., 1995	Atlantic Ocean	01/06/1992 - 31/07/1994	X	X	X			
J	Gustafsson et al., 1997b	Atlantic Ocean	01/06/1993 - 31/05/1994	X		X	X		
J	Langone et al., 1997	Southern Ocean	07/12/1994 - 13/12/1994	X	X	X			
J	Moran et al., 1997	Arctic Ocean	01/08/1994 - 30/09/1994	X	X	X	X		X
J	Rutgers van der Loeff et al., 1997 & Friedrich and Rutgers van der Loeff, 2002	Southern Ocean	02/10/1992 - 22/11/1992		X	X	X		X
J	Buesseler et al., 1998	Indian Ocean	09/01/1995 - 12/09/1995		X	X	X		X
J	Gustafsson et al., 1998	Atlantic Ocean	26/09/1993 - 22/05/1994	X		X			
J	Wei and Hung, 1998	Pacific Ocean	01/11/1991 - 30/11/1991		X	X			
J	Kersten et al., 1998	Atlantic Ocean	02/02/1994 - 09/11/1994		X	X			
J	Charette et al., 1999	Pacific Ocean	01/02/1996 - 28/02/1997	X	X	X	X	X	
J	Charette and Moran, 1999	Atlantic Ocean	15/05/1996 - 11/06/1996	X	X	X	X		
J	Santschi et al., 1999	Atlantic Ocean	17/05/1993 - 12/07/1994	X	X	X	X		
J	Smoak et al., 1999	Pacific Ocean	16/10/1993 - 31/03/1996				X		
J	Benitez-Nelson et al., 2000	Atlantic Ocean	01/03/1997 - 31/08/1997		X	X	X		
J	Buesseler et al., 2000	Atlantic Ocean	06/10/1997 - 06/10/1997				X	X	X
J	Charette and Buesseler, 2000	Southern Ocean	09/02/1999 - 23/02/1999	X	X	X	X		X

https://doi.org/10.5194/essd-2021-259
Preprint. Discussion started: 1 October 2021
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J	Cochran et al., 2000	Southern Ocean	08/10/1996 - 04/05/1997	X		X	X		X	
J	Gulin, 2000	Atlantic Ocean	02/11/1998 - 11/06/1999	X	X	X				
J	Hall et al., 2000	Atlantic Ocean	11/06/1997 - 14/06/1997	X	X	X	X			X
J	Moran and Smith, 2000	Arctic Ocean	01/08/1995 - 30/09/1995	X	X	X	X		X	
J	Benitez-Nelson et al., 2001a	Pacific Ocean	01/04/1999 - 31/03/2000	X			X	X		
J	Buesseler et al., 2001b	Southern Ocean	24/10/1997 - 15/03/1998	X	X	X	X		X	
J	Pinghe et al., 2001	Pacific Ocean	01/04/1999 - 01/05/1999	X	X	X	X			
J	Charette et al., 2001	Atlantic Ocean	19/03/1995 - 31/08/1997	X	X	X	X	X		
J	Dai and Benitez-Nelson, 2001	Atlantic Ocean	06/07/1996 - 12/07/1997		X	X	X			
J	Kim and Church, 2001	Atlantic Ocean	06/10/1996 - 20/08/1997	X	X	X				
J	Hernes et al., 2001	Pacific Ocean	07/02/1992 - 13/09/1992				X	X		
J	Kim and Church, 2001	Atlantic Ocean	06/10/1996 - 20/08/1997	X	X	X				
J	Porcelli et al., 2001	Atlantic Ocean	01/06/1995 - 1995/06/31	X	X	X				
J	Turnewitsch and Springer, 2001	Atlantic Ocean	01/07/1997 - 28/02/1998	X	X	X				
R	Turnewitsch, 2001	Indian Ocean	01/02/1998 - 31/03/1998		X	X				
J	Amiel et al., 2002	Atlantic Ocean	01/05/1998 - 30/09/1999	X	X	X	X	X		
J	Cai et al., 2002	Pacific Ocean	15/11/1997 - 15/11/1997	X	X	X	X			
J	Coppola et al., 2002	Arctic Ocean	07/07/1999 - 07/07/1999	X	X	X	X	X		
J	Foster and Shimmield, 2002	Atlantic Ocean	19/06/1999 - 27/06/1999	X	X	X	X			
J	Guo et al., 2002	Atlantic Ocean	05/07/2000 - 05/07/2000	X	X	X	X			
J	Rutgers van der Loeff et al., 2002a	Southern Ocean	30/12/1995 - 18/01/1996	X	X	X	X			
J	Rutgers van der Loeff et al., 2002b	Arctic Ocean	26/06/1997 - 11/08/1997	X	X	X				
J	Schmidt et al., 2002b & Schmidt et al., 2009	Mediterranean Sea	09/02/1994 - 29/05/1995	X	X	X	X	X		
J	Schmidt et al., 2002a	Atlantic Ocean	01/06/1997 - 31/01/1998	X	X	X	X			X
J	Somayajulu et al., 2002	Bay of Bengal	16/12/1991 - 23/12/1991	X						
J	Usbeck et al., 2002	Southern Ocean	20/03/1999 - 06/05/1999	X	X	X				

https://doi.org/10.5194/essd-2021-259
Preprint. Discussion started: 1 October 2021
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J	Baskaran et al., 2003	Arctic Ocean	24/07/1998 - 10/10/1998	X	X	X	X		
J	Chen et al., 2003	Pacific & Arctic Oceans	01/08/1999 - 31/08/1999	X	X	X	X		
J	Moran et al., 2003	Atlantic Ocean	01/07/1999 - 31/07/1999	X	X	X	X		
J	Radakovitch et al., 2003	Mediterranean Sea	06/03/1997 - 12/09/1997	X	X	X	X		
R	Rutgers van der Loeff and Vöge, 2003	Southern Ocean	24/10/2000 - 29/11/2000	X	X	X			
J	Santschi et al., 2003	Atlantic Ocean	01/05/2001 - 31/05/2001	X	X	X	X	X	
J	Sweeney et al., 2003	Atlantic Ocean	01/03/1993 - 30/09/1995				X	X	
J	Savoye et al., 2004 & Buesseler et al., 2006	Southern Ocean	03/11/2001 - 05/12/2001	X		X	X		X
J	Gustafsson et al., 2004	Atlantic Ocean	17/01/1998 - 15/08/2000	X		X			
J	Hung et al., 2004	Atlantic Ocean	07/07/2000 - 21/05/2001	X	X	X	X		
J	Kawakami et al., 2004 & Kawakami, 2009 & Yang et al., 2004	Pacific Ocean	12/11/1997 - 04/06/2000		X	X	X		
J	Smith et al., 2004	Atlantic Ocean	15/06/1998 - 15/07/1998		X	X			
J	Smoak et al., 2004	Atlantic Ocean	15/11/1995 - 30/04/1996					X	
J	Trimble et al., 2004	Arctic Ocean	01/08/2000 - 31/08/2000		X	X			
J	Waples, 2004	Lake Michigan	15/02/1999 - 18/10/1999		X	X			
J	Aono et al., 2005	Pacific Ocean	07/07/2001 - 31/07/2001	X	X	X	X	X	X
J	Buesseler et al., 2005	Southern Ocean	30/01/2002 - 20/02/2002	X		X	X		
J	Coppola et al., 2005 & Coppola et al., 2006	Southern Ocean	18/01/1999 - 15/02/1999	X	X	X	X		
J	Ma et al., 2005	Arctic Ocean	07/01/2003 - 30/09/2003	X		X	X		
J	Moran et al., 2005 & Lepore and Moran, 2007	Arctic Ocean	06/05/2002 - 26/08/2002	X	X	X	X		X
J	Trimble and Baskaran, 2005	Arctic Ocean	01/08/2000 - 31/08/2000	X	X	X	X		X
J	Cai et al., 2006b	Pacific Ocean	21/02/2004 - 28/02/2004	X	X	X			
J	Cai et al., 2006c	Pacific Ocean	04/05/2005 - 09/05/2005	X	X	X	X		
J	Gustafsson et al., 2006	Atlantic Ocean	15/08/2000 - 31/03/2001				X	X	

https://doi.org/10.5194/essd-2021-259
Preprint. Discussion started: 1 October 2021
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J	Rodriguez y Baena et al., 2006, 2008	Southern Ocean	17/11/2003 - 18/01/2004	X			X				
J	Schmidt, 2006	Atlantic Ocean	26/12/1988 - 08/09/1989	X	X	X	X				
J	Speicher et al., 2006	Mediterranean Sea	02/05/2004 - 16/03/2004	X	X	X	X				
J	Thomalla et al., 2006, 2008	Atlantic Ocean	02/05/2004 - 26/05/2004	X			X				
J	Giuliani et al., 2007	Mediterranean Sea	16/03/1997 - 07/04/1997	X	X	X	X				
J	Hung and Gong, 2007	Pacific Ocean	18/08/2006 - 18/08/2006	X	X	X	X				
J	Kawakami and Honda, 2007	Pacific Ocean	16/10/2002 - 21/08/2004		X	X	X				
J	Lalande et al., 2007 & Lepore et al., 2007 & Lepore and Moran, 2007	Arctic Ocean	24/05/2004 - 25/08/2004	X	X	X	X	X	X		
J	Morris et al., 2007	Southern Ocean	10/11/2004 - 14/01/2005	X			X				
R	Rutgers van der Loeff, 2007a	Atlantic Ocean	21/10/2005 - 13/11/2005	X	X	X					
R	Rugters van der Loeff, 2007b	Southern Ocean	16/01/2006 - 30/01/2006	X	X	X					X
J	Stewart et al., 2007	Mediterranean Sea	04/03/2003 - 30/06/2003	X			X	X			
T	Thomalla, 2007	Atlantic Ocean	17/05/2003 - 04/10/2003		X	X	X				
J	Buesseler et al., 2008	Atlantic Ocean	24/06/2004 - 25/08/2005	X		X	X	X	X		
J	Cai et al., 2008	Pacific Ocean	04/04/2004 - 04/05/2004	X		X	X				
J	Cochran and Kadko, 2008	Arctic Ocean	01/09/2002 - 31/07/2004	X	X	X	X				
J	Lalande et al., 2008	Arctic Ocean	10/07/2003 - 31/05/2005	X	X	X	X	X			
J	Lamborg et al., 2008b	Pacific Ocean	23/06/2004 - 09/08/2005				X				
J	Lampitt et al., 2008	Atlantic Ocean	12/07/2003 - 06/07/2007	X			X	X			
J	Chen et al., 2008	Pacific Ocean	01/07/2000 - 30/11/2002	X	X	X	X				
J	Maiti et al., 2008	Pacific Ocean	10/03/2005 - 28/03/2005	X			X	X	X		
J	Savoye et al., 2008	Southern Ocean	23/01/2005 - 12/02/2005	X			X	X	X		
J	Brew et al., 2009 & Stewart et al., 2011	Atlantic Ocean	09/05/2006 - 24/03/2007	X		X	X	X			

https://doi.org/10.5194/essd-2021-259
Preprint. Discussion started: 1 October 2021
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J	Buesseler et al., 2009	Pacific Ocean	23/06/2004 - 16/08/2005	X		X	X	X	X
R	Charette, 2009	Southern Ocean	16/01/2006 - 06/08/2006	X					
J	Cochran et al., 2009	Mediterranean Sea	10/03/1999 - 30/04/2005	X	X	X			
J	Lepore et al., 2009	Atlantic Ocean	06/05/2005 - 03/12/2004	X	X	X	X	X	
J	Schmidt et al., 2009	Mediterranean Sea	03/09/2004 - 24/09/2004	X	X	X	X	X	
J	Szlosek et al., 2009	Mediterranean Sea	06/03/2003 - 01/05/2005				X	X	
J	Wei et al., 2009	Pacific Ocean	06/03/2006 - 08/03/2006		X	X	X		
J	Buesseler et al., 2010	Southern Ocean	09/01/2009 - 09/01/2009	X		X	X	X	
J	Cai et al., 2010	Arctic Ocean	30/07/2007 - 23/09/2007	X		X	X		
J	Hung and Gong, 2010	Pacific Ocean	03/12/2008 - 04/12/2008				X	X	
J	Hung et al., 2010	Atlantic & Pacific Oceans	01/08/2005 - 16/06/2009	X	X	X	X	X	
J	Kawakami et al., 2010	Pacific Ocean	26/09/2005 - 17/10/2005		X	X	X	X	
J	Sanders et al., 2010	Atlantic Ocean	24/07/2007 - 23/08/2007	X		X	X		X
J	Yu et al., 2010	Arctic Ocean	30/07/2003 - 12/09/2003	X	X	X	X		
J	Evangeliou et al., 2011	Mediterranean Sea	01/07/2008 - 17/01/2009	X	X	X	X		
J	Jacquet et al., 2011	Southern Ocean	17/01/2007 - 20/02/2007	X		X	X	X	
J	Kim et al., 2011	Pacific Ocean	04/04/2007 - 23/02/2008	X			X		
J	Martin et al., 2011	Atlantic Ocean	01/05/2008 - 21/05/2008	X			X	X	X
J	Rutgers van der Loeff et al., 2011	Southern Ocean	13/02/2008 - 11/04/2008	X		X	X		
J	Shaw et al., 2011	Southern Ocean	15/03/2009 - 15/04/2009	X			X	X	
J	Stukel et al., 2011	Pacific Ocean	11/05/2006 - 05/06/2006	X			X		
J	Wei et al., 2011	Pacific Ocean	01/10/2006 - 31/12/2008	X	X	X	X	X	
J	Xu et al., 2011	Atlantic Ocean	30/04/2006 - 05/05/2006			X	X	X	
J	Gustafsson and Andersson, 2012	Arctic Ocean	13/07/2001 - 31/07/2001	X	X	X	X		
J	Hung et al., 2012	Pacific Ocean	02/08/2008 - 12/12/2008				X	X	
R	Kawakami, 2012	Pacific Ocean	09/09/2007 - 28/10/2008		X	X	X	X	X
J	Moran et al., 2012	Pacific Ocean	29/03/2008 - 31/07/2008	X			X	X	
J	Yu et al., 2012	Arctic Ocean	04/08/2008 - 08/09/2008	X	X	X	X		
J	Zhou et al., 2012	Southern Ocean	24/05/2008 - 08/06/2008	X		X	X		
J	Baumann et al., 2013	Pacific Ocean	29/03/2008 - 15/07/2010	X			X	X	

https://doi.org/10.5194/essd-2021-259
Preprint. Discussion started: 1 October 2021
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J	Evangeliou et al., 2013	Mediterranean Sea	01/01/2010 - 10/01/2010	X	X	X	X			
J	Haskell et al., 2013	Pacific Ocean	01/02/2010 - 03/04/2011	X			X	X		
R	JGOFS India2013	Indian Ocean	09/02/1997 - 11/02/1997		X					
J	Le Moigne et al., 2013a	Atlantic Ocean	13/07/2009 - 08/08/2009	X		X	X			
T	Luo, 2013 & Luo et al., 2014	Pacific Ocean	01/06/2010 - 30/06/2010	X	X	X	X	X		
J	Martin et al., 2013	Southern Ocean	12/01/2009 - 06/03/2009	X	X	X	X	X		X
T	Owens 2013	Atlantic Ocean	10/09/2010 - 05/10/2010	X		X	X		X	
T	Owens 2013	Southern Ocean	30/12/2009 - 07/02/2010	X		X	X	X	X	X
T	Owens 2013	Southern Ocean	16/03/2010 - 01/05/2010	X		X	X	X	X	X
J	Planchon et al., 2013	Southern Ocean	21/02/2008 - 14/03/2008	X		X	X			
J	Schmidt et al., 2013	Atlantic Ocean	01/05/2002 - 31/05/2002	X	X	X	X			
J	Stukel et al., 2013	Pacific Ocean	04/04/2007 - 28/10/2008	X			X	X		
J	Zhou et al., 2013	Pacific Ocean	01/08/2007 - 31/08/2007	X		X	X			
J	Le Moigne et al., 2014	Atlantic Ocean	19/10/2010 - 24/01/2012	X		X	X		X	
J	Luo et al., 2014	Pacific Ocean	10/03/2009 - 09/02/2011	X	X	X	X		X	
T	Pabortsava, 2014	Atlantic Ocean	07/02/2011 - 07/02/2011	X		X	X		X	
J	Cai et al., 2015	Pacific Ocean	18/07/2009 - 24/05/2011	X		X	X			
J	Estapa et al., 2015	Atlantic Ocean	30/09/2011 - 12/02/2012	X						X
J	Kawakami et al., 2015	Pacific Ocean	03/06/2006 - 17/07/2006		X	X	X	X		
J	Le Moigne et al., 2015	Arctic Ocean	05/06/2012 - 01/07/2012	X		X	X			
J	Owens et al., 2015	Atlantic Ocean	15/10/2010 - 07/04/2011	X		X	X			X
J	Planchon et al., 2015	Southern Ocean	20/10/2011 - 18/11/2011	X		X	X	X		
J	Puigcorbé et al., 2015	Pacific Ocean	10/07/2008 - 07/08/2008	X		X	X	X	X	
R	Roca-Martí, et al.., 2015	Southern Ocean	26/01/2012 - 02/03/2012	X		X	X		X	
J	Rosengard et al., 2015	Atlantic & Indian Oceans	14/01/2012 - 20/03/2012	X		X	X			
J	Stukel et al., 2015, 2016, 2017	Pacific Ocean	22/04/2009 - 23/07/2010	X			X	X		
J	Ceballos-Romero et al., 2016	Atlantic Ocean	01/05/2010 - 07/08/2010	X		X	X	X		
J	Haskell et al., 2016	Pacific Ocean	16/01/2013 - 19/06/2014	X			X	X		

https://doi.org/10.5194/essd-2021-259
Preprint. Discussion started: 1 October 2021
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J	Le Moigne et al., 2016	Southern Ocean	13/01/2013 - 05/02/2013	X		X	X			
J	Maiti et al., 2016	Atlantic Ocean	01/03/2012 - 31/03/2013		X		X	X		
J	Roca-Martí et al., 2016	Arctic Ocean	03/08/2012 - 08/10/2012	X		X	X		X	
J	Turnewitsch et al., 2016	Atlantic Ocean	28/09/2009 - 19/10/2009	X		X	X			
J	Zhou et al., 2016	Atlantic Ocean	05/06/2008 - 30/09/2009		X	X	X		X	
J	Anand et al., 2017	Indian Ocean	16/03/2014 - 19/04/2014	X		X	X			
J	Puigcorbé et al., 2017a	Atlantic Ocean	02/05/2010 - 04/07/2010	X			X			
J	Puigcorbé et al., 2017b	Southern Ocean	07/01/2012 - 11/03/2012	X		X	X	X	X	
J	Roca-Martí et al., 2017	Southern Ocean	07/01/2012 - 11/03/2012	X		X	X	X	X	
J	Anand et al., 2018b	Indian Ocean	24/10/2013 - 29/01/2014	X			X			
J	Anand et al., 2018a	Indian Ocean	09/05/2014 - 28/05/2014	X		X	X			
J	Black et al., 2018	Pacific Ocean	29/10/2013 - 19/12/2013	X		X	X			X
J	Lemaitre et al., 2018	Atlantic Ocean	20/05/2014 - 26/06/2014	X		X	X			
J	Roca-Martí et al., 2018	Mediterranean Sea	04/05/2013 - 30/05/2013	X						
J	Stukel et al., 2019	Pacific Ocean	09/08/2014 - 11/05/2016	X			X	X		
R	Stukel, 2019	Pacific Ocean	01/06/2017 - 29/06/2017	X						
J	Umhau et al., 2019	Pacific Ocean	01/02/2014 - 30/09/2015	X		X	X	X	X	
J	Alkalay et al., 2020	Mediterranean Sea	25/05/2017 - 03/06/2018	X			X	X		
J	Buesseler et al., 2020a	Pacific Ocean	14/08/2018 - 10/09/2018	X		X	X	X		
J	Xie et al., 2020	Pacific Ocean	13/04/2017 - 23/07/2017	X						
P	Pers. comm. from K. Buesseler	Southern Ocean	06/01/2009 - 30/01/2009	X		X	X		X	
P	Pers. comm. from K. Buesseler	Southern Ocean	20/02/2009 - 14/03/2009	X						
P	Pers. comm. from E. Ceballos-Romero 2021	Southern Ocean	01/12/2013 - 12/12/2013	X		X	X			X
P	Pers. comm. from Gasser	Pacific Ocean	27/10/2004 - 11/12/2004	X	X	X				
P	Pers. comm. from J. Kenyon	Pacific Ocean	25/09/2018 - 23/10/2018	X		X	X		X	
P	Pers. comm. from K. Kenyon	Pacific Ocean	26/10/2018 - 24/11/2018	X		X	X		X	

https://doi.org/10.5194/essd-2021-259
Preprint. Discussion started: 1 October 2021
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P	Pers. comm. from S. Owens	Atlantic Ocean	08/11/2010 - 26/11/2010	X	X				X
P	Pers. comm. from K. Pabortsava	Atlantic Ocean	11/08/2011 - 07/09/2011	X		X	X	X	
P	Pers. comm. from V. Puigcorbe	Mediterranean Sea	08/03/2009 - 20/09/2009	X		X	X		

815

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Table 2 Major Ocean programs identified in the 50 years compiled and significant activities within these programs with ^{234}Th samplings chronologically ordered. Activities are categorized as follows: i) cruises (C), ii) experiments (E), iii) Long-Term Time-Series station (TSS), iv) Process Study, and v) subprogram (SP). Programs and activities are detailed by era, as indicated with a cross.

Ocean programs						Eras			
#	Acronym	Activity	Name	Type	Dates	1	2	3	4
1	GEOSECS		Geochemical Ocean Sections Study		1972-1978	x			
2	LTER		Long Term Ecological Research (LTER)		1980-present				
		PAL-LTER	Palmer Antartica Long Term Ecologycal Research	SP	1990-present				x
		CCE-LTER	California Current Ecosystem Long Term Ecologycal Research	SP	2004-present				x
3	JGOFS		Joint Global Ocean Flux Study		1987-2003				
		DYFAMED	DYnamique des Flux de mAtière en MEDiterranée	TSS	1897-present	x	x	x	
		BATS	Bermuda Atlantic Time-series Study	TSS	1988-present		x	x	x
		HOT	Hawaii Ocean Time-Series	TSS	1988-present			x	x
		MEDATLANTE	MEDATLANTE	SP	1989-1990			x	
		NABE	North Atlantic Bloom Experiment	PS	1989		x		
		OSP	Station PAPA Ocean Weather Station "P"	TSS	1990*-present		x		x
		EqPac	Equatorial Pacific Project	PS	1992		x		
		ANTARES	ANTArctic RESearch	SP	1992-1998			x	
		Arabian Sea	Arabian Sea Process Study	PS	1994-1996		x		
		DYNAPOC	DYNAPROC	C	1995			x	
		AESOPS	Antarctic Environment and Southern Ocean Process Study	PS	1996-1998		x		
		KNOT	Kyodo North Pacific Ocean Time Series	TSS	1998-2000			x	
		SEATS	South-East Asia Time Series	TSS	1999-present		x	x	x
		SOIREE	Southern Ocean Iron Release Experiment	E	1999		x		
4	OMEX		Ocean Margins Exchange Program (I & II)		1993-2000		x	x	
5	CARIACO		CArbon Retention In A Colored Ocean time series program		1995-2017			x	
6	SBI		Shelf-Basin Interactions (SBI) Phase II field program		1998-2009			x	
7	HiLATS		High Latitude Time Series Observatory project		2001			x	
8	MedFlux		MedFlux		2003-2006			x	
9	AMT		Atlantic Meridional Transect (AMT) programme		2007-2012			x	
10	BEST		BEST-BSIERP field program		2007-2013				x

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1	CHOICE	Carbon Cycling in the China Seas: Budget, Controls and			
1	-C	Ocean Acidification	2009-2011	x	x
1					
2	UKOA	UK's Ocean Acidification	2010-2016		x
1			2010-		
3	GEOTRACES	GEOTRACES	present	x	x
1	EXPOR		2017-		
4	TS	NASA EXport Processes in the Ocean from RemoTe Sensing	present		x

820

*Oservations at Station Papa (former Ocean Station Peter and referred to as OSP or Ocean Station “P”) started in 1949, although the larger surveys became a focus of activities in support of JGOFS during the 1990s (Freeland, 2007).

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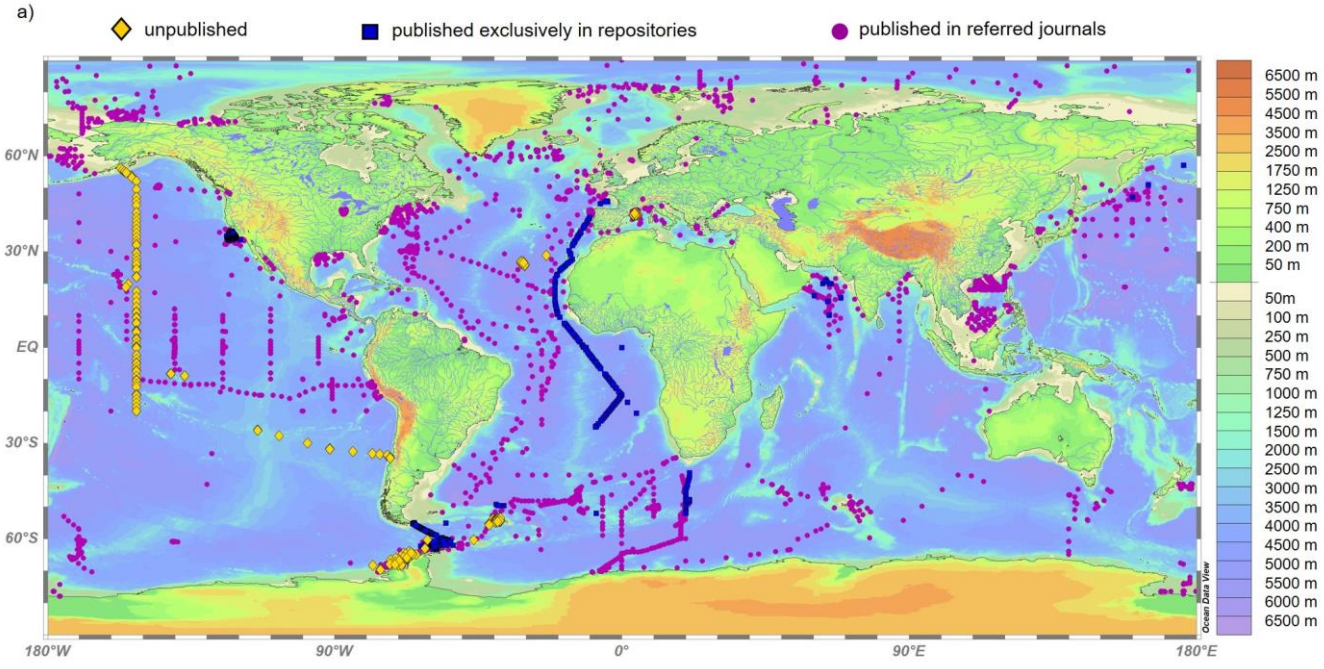
825 **Figures**

Figure 1. (a) Map showing the distribution of sampling stations catalogued as i) unpublished (yellow diamonds), ii) published exclusively in repositories (blue square), and iii) published in referred journals (magenta circles). (b) Map showing data density by sampling location. (c) Map showing long-term, high-frequency time-series stations (LTTS) either i) "operational" (blue diamond), ii) "registered" (for future operation (yellow diamond), iii) "inactive" (red
830 **asterisk) or iv) "close" (black cross) (source: https://www.ocean-ops.org) and those locations including ^{234}Th sampling that match a TTS station (light blue circles).**



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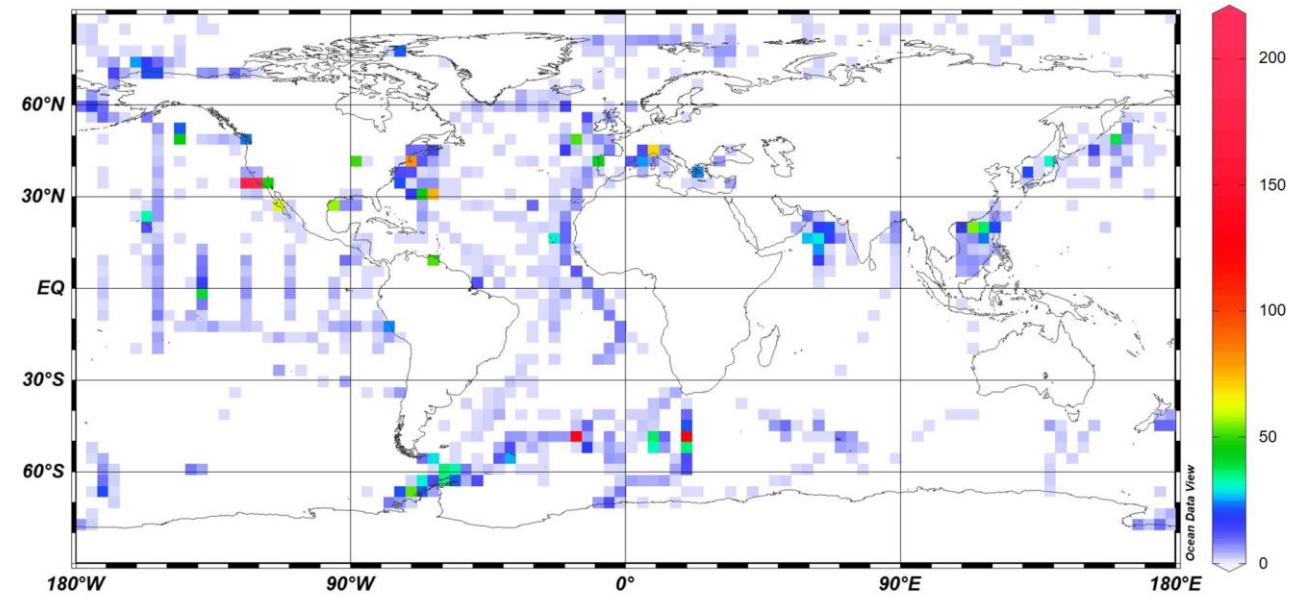
b)



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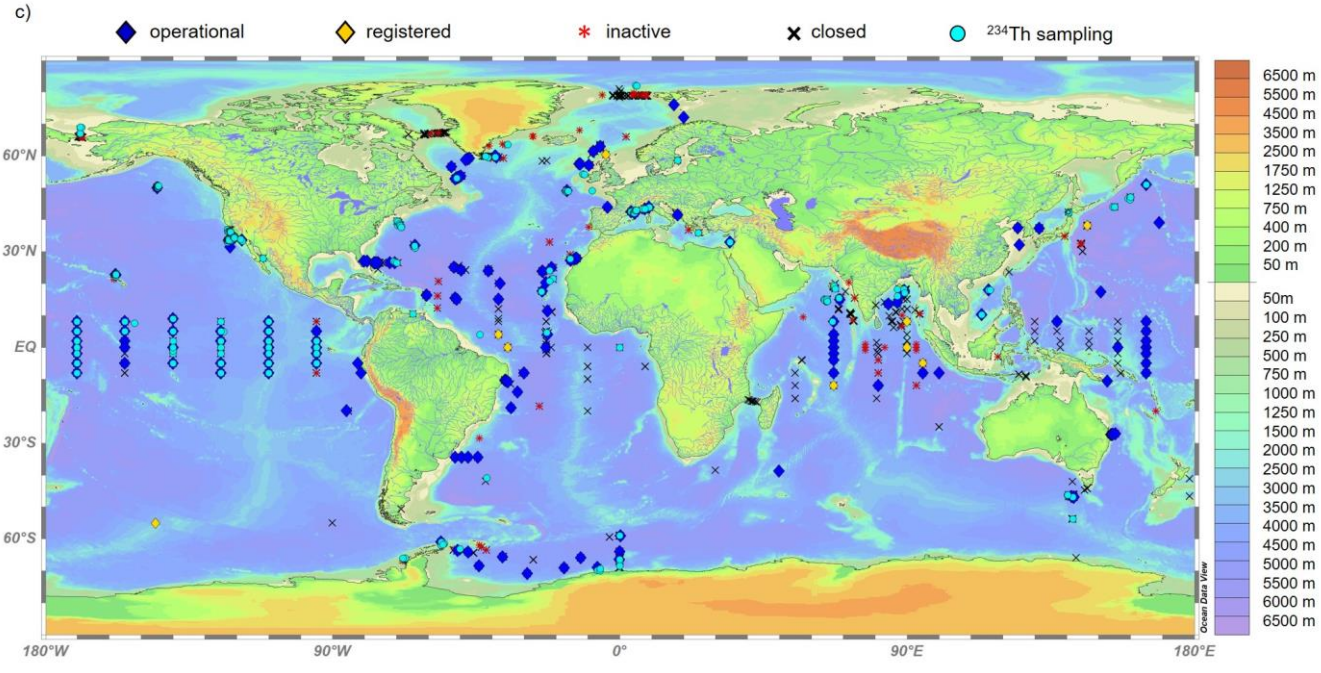

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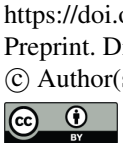

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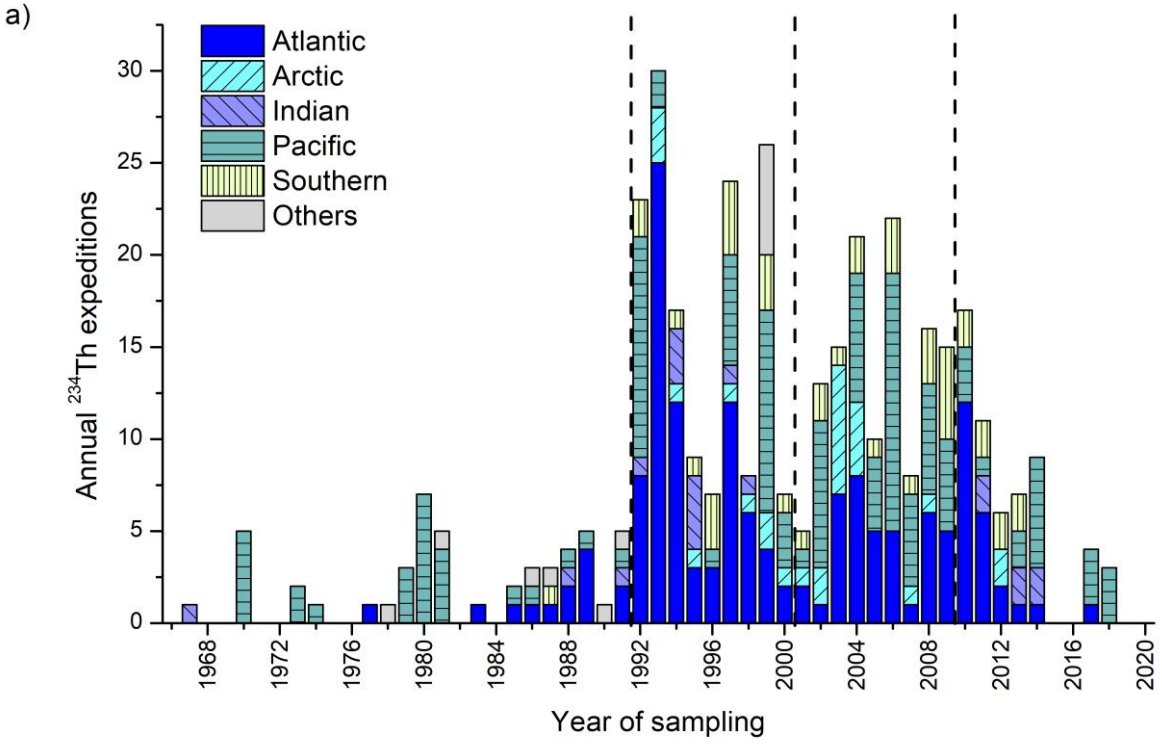
Figure 2. (a) Time distribution of annual field surveys with ^{234}Th measurements since the first reported sampling in 1967 separated by Ocean. (b) Equivalent figure for ^{234}Th publications (either in referred journals or repositories). Note that Oceans are distinguished as i) Atlantic, ii) Arctic, iii) Indian, iv) Pacific and v) Southern, with and additional category for vi) "Others", which includes rivers, lakes, and bays. (c) Histogram of ^{234}Th data points sampled since 1967. separated by data type as follows: i) ^{238}U, ii) total ^{234}Th, iii) dissolved ^{234}Th, iv) particulate ^{234}Th, v) POC:^{234}Th ratios and vi) PON:^{234}Th ratios. Dotted lines indicate tipping points in the ^{234}Th timeline.



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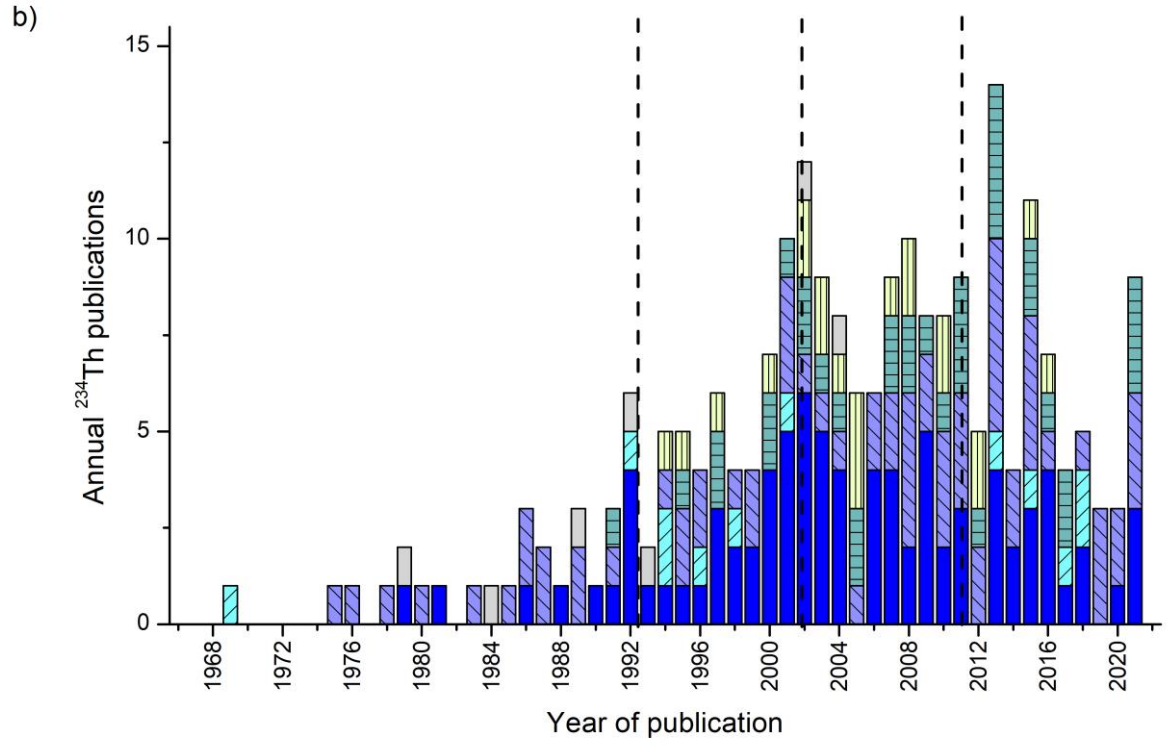

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c)

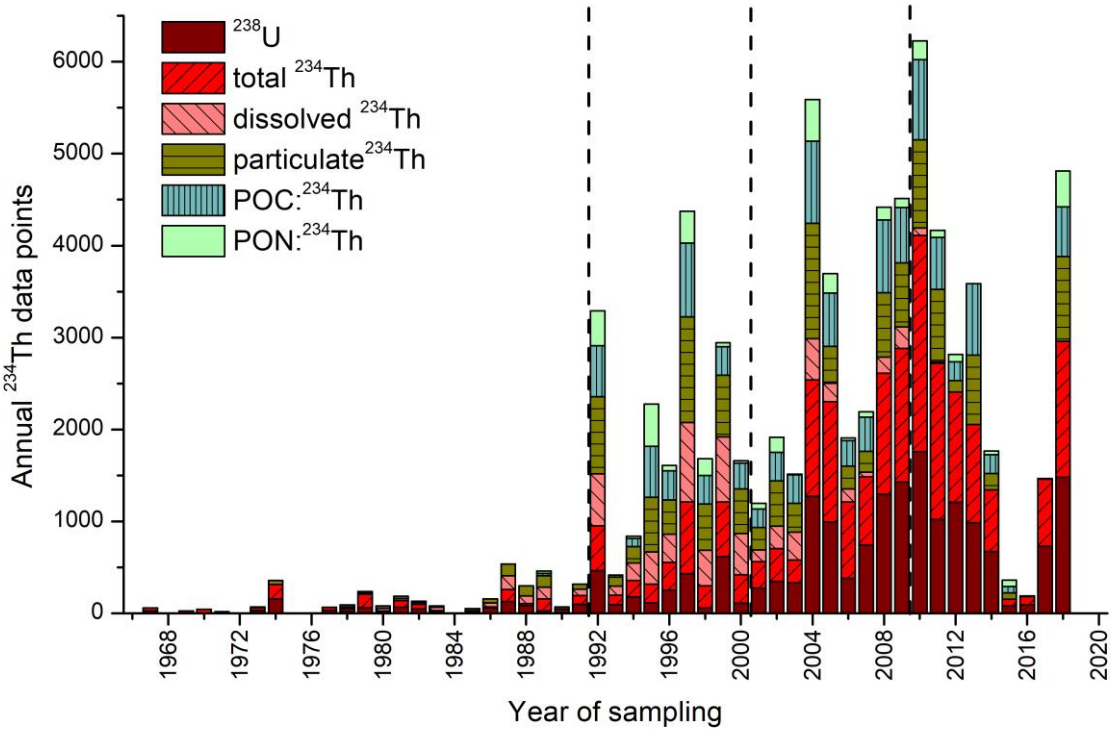

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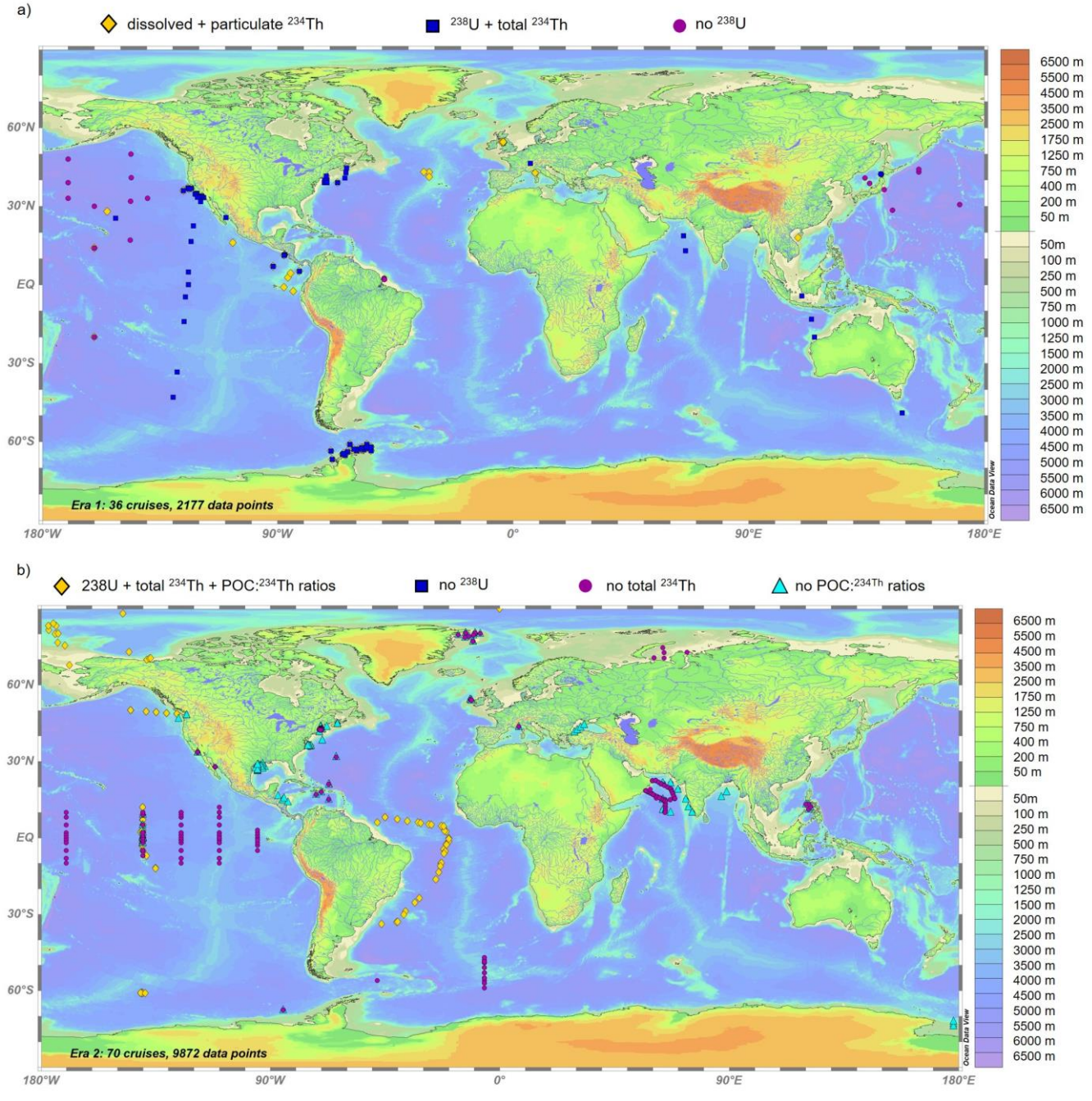
Figure 3. Map showing the distribution of sampling stations (a) during era 1 distinguished by the by the type of data available as follows: i) dissolved plus particulate ^{234}Th (yellow diamonds), ii) ^{238} U plus total ^{234}Th (magenta dots), iii) no ^{238}U (dark blue squares); and for eras 2 (b), 3 (c) and 4 (d) distinguished by the type of data available as follows: i) total ^{238}U plus total ^{234}Th plus POC:^{234}Th ratios (yellow diamonds), ii) ^{238}U not sampled (dark blue squares), iii) total ^{234}Th not sampled (light blue triangles), and iv) POC:^{234}Th ratios not sampled (magenta dots). Therefore, only locations that fall into categories i) and ii) can be used to estimate POC fluxes.



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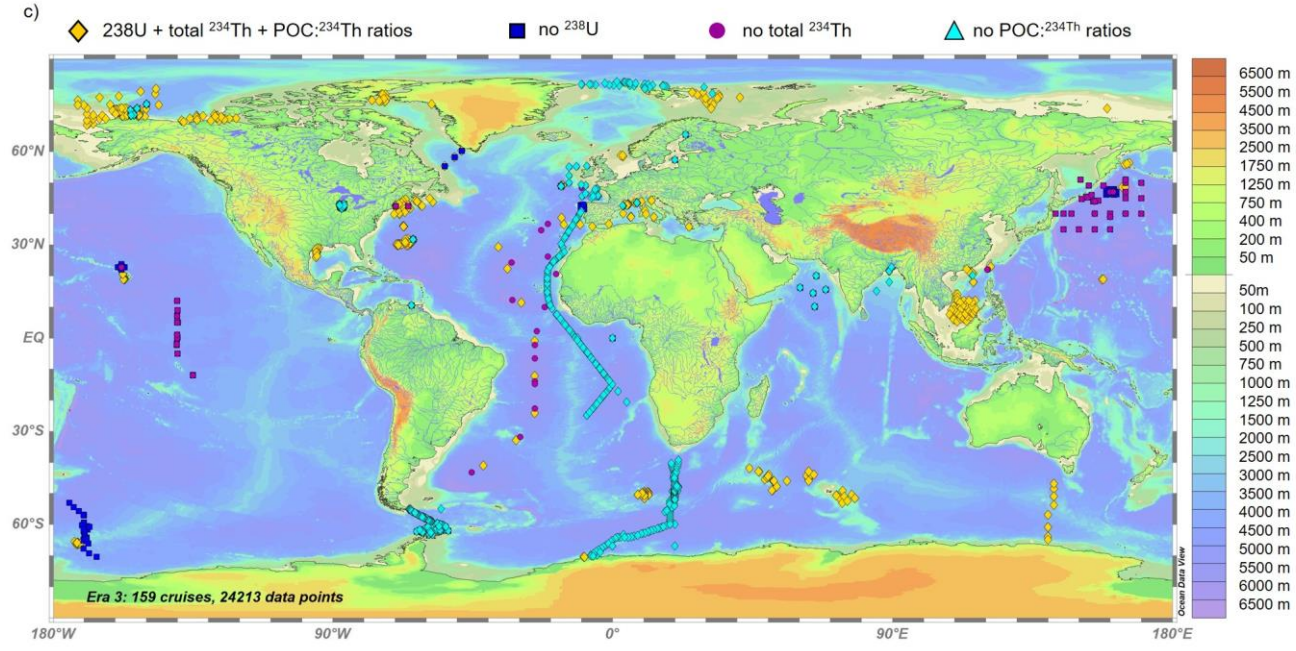

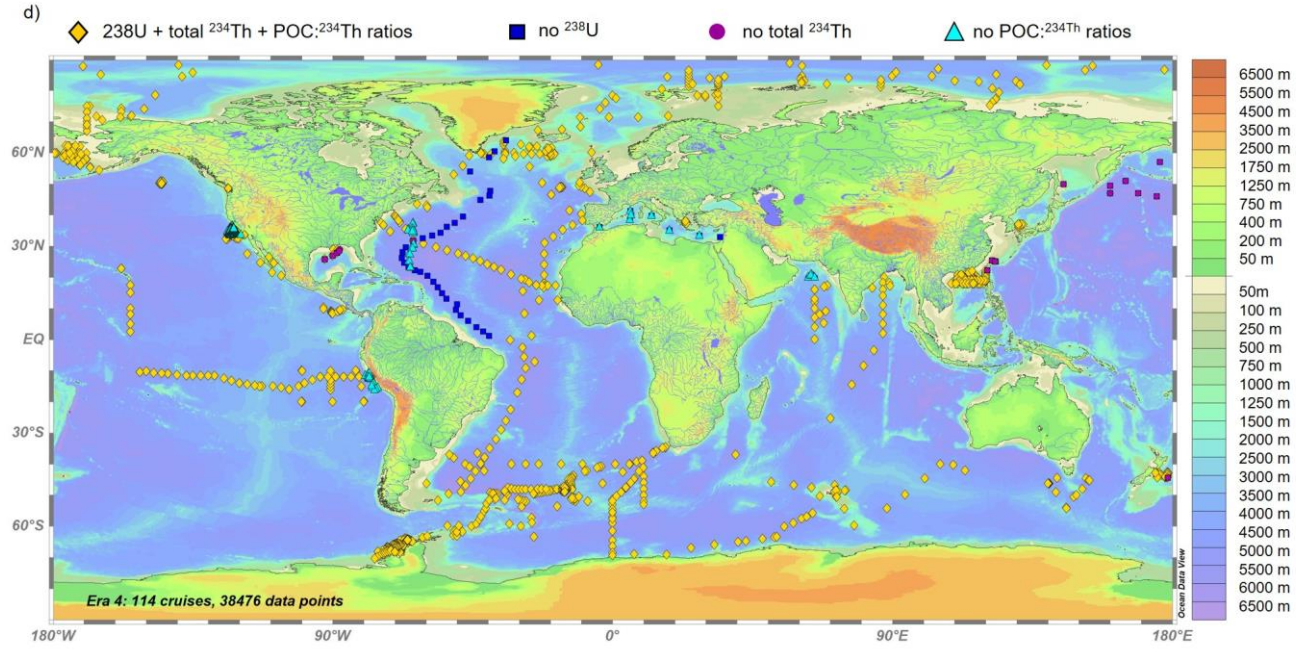

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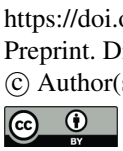

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Figure 4. Seasonal distribution of annual field expeditions with ^{234}Th sampling within eras 1 (a), 2 (b), 3 (c) and 4 (d) separated by hemisphere as follows: i) Northern Hemisphere (HN), ii) Southern Hemisphere (SH), and iii) crossing the Equator and sampling in both hemispheres (both).

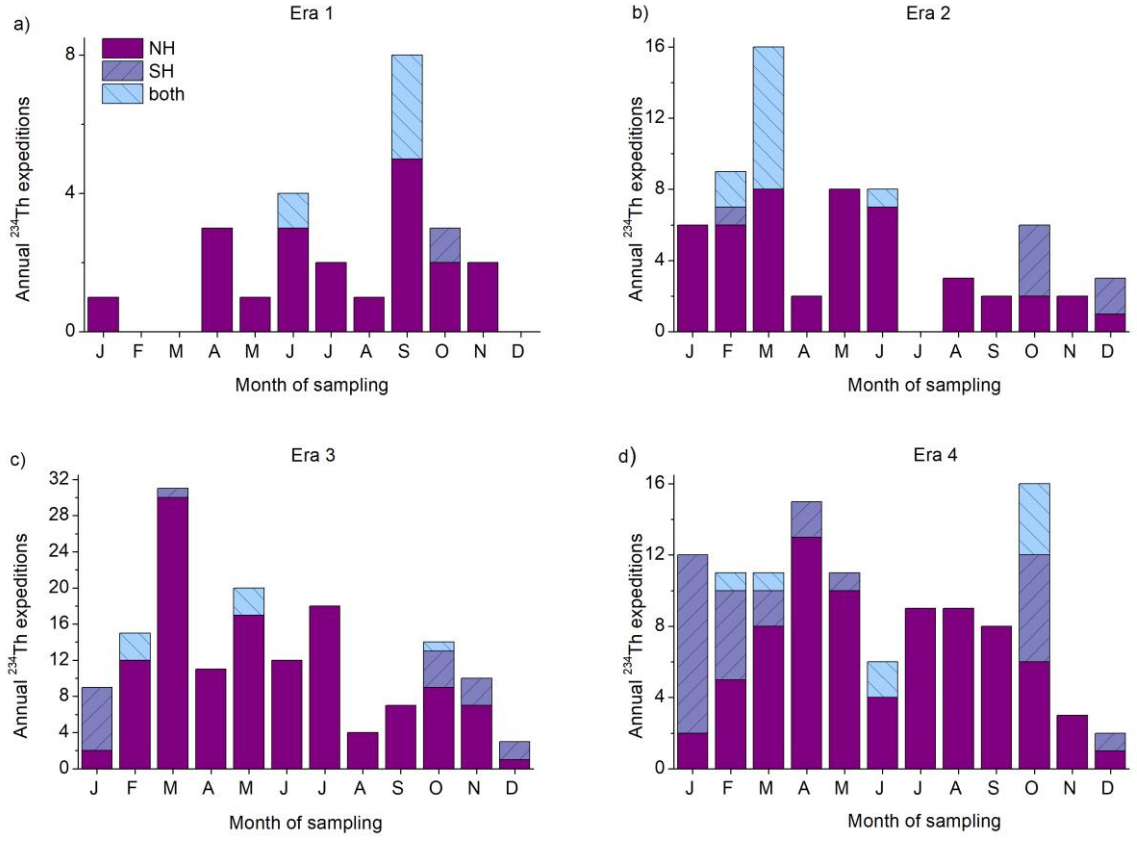

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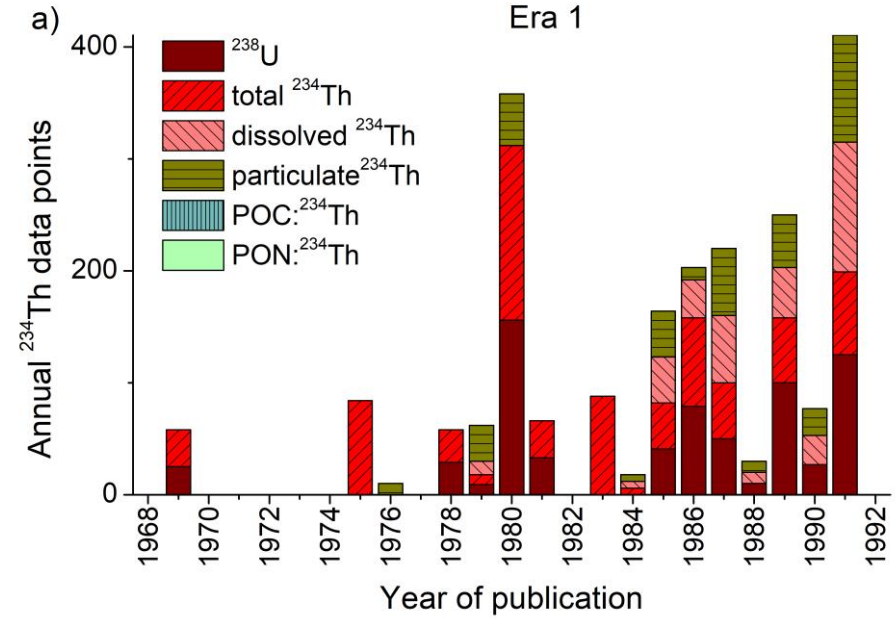
Figure 5. Histogram of data points published within eras 1 (a), 2 (b), 3 (c) and 4 (d) separated by data type as follows: i) ^{238}U, ii) total ^{234}Th, iii) dissolved ^{234}Th, iv) particulate ^{234}Th, v) POC:^{234}Th ratios and vi) PON:^{234}Th ratios.



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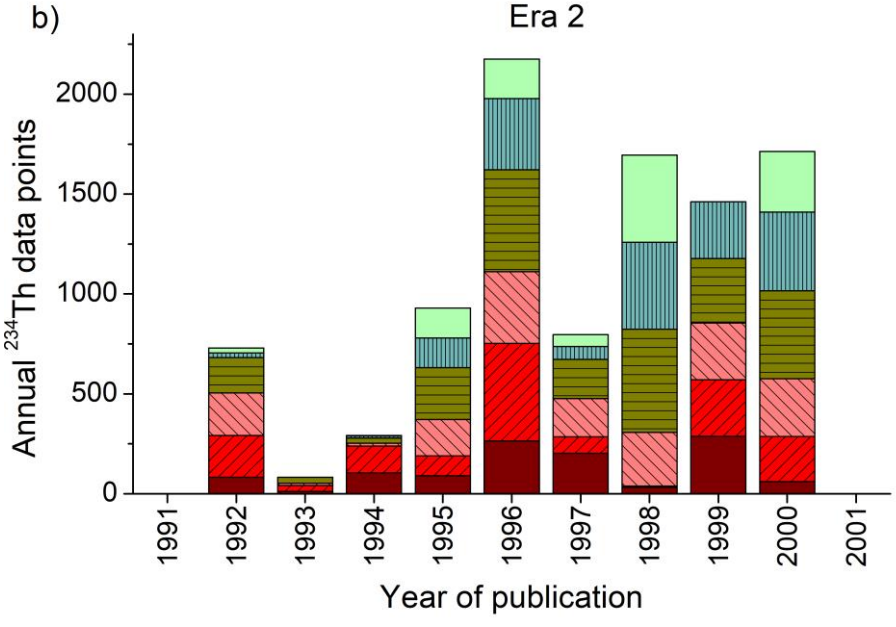

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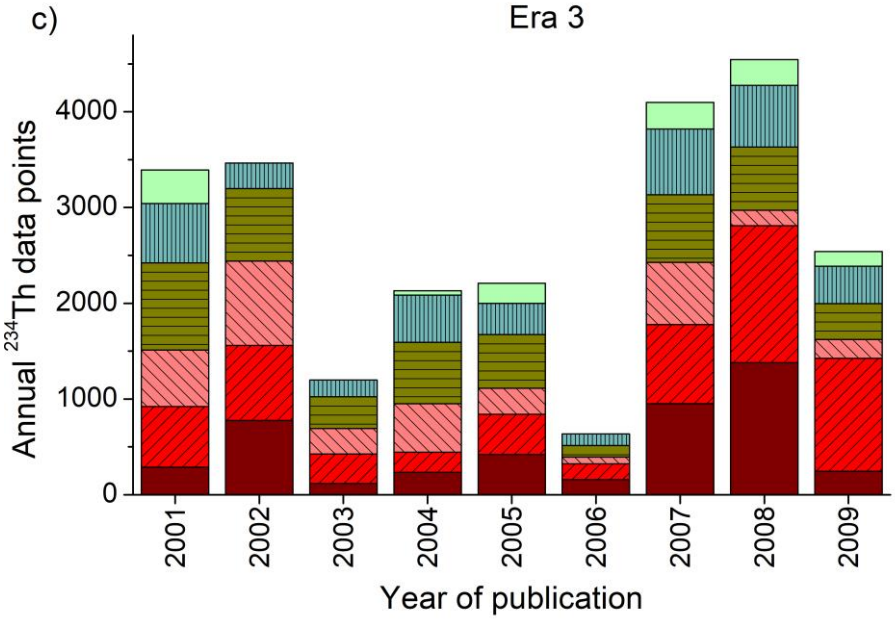

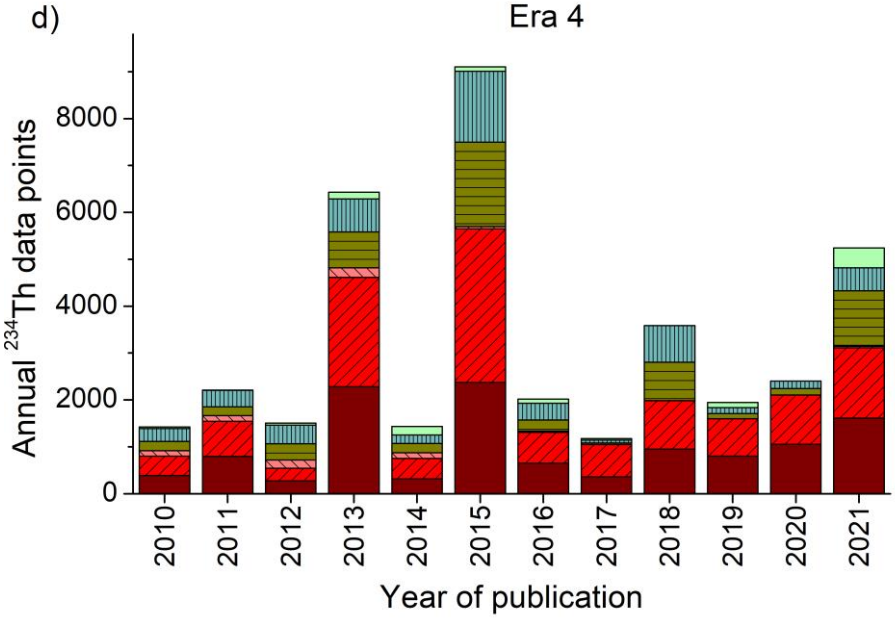

915

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