

The author and the other reviewers have addressed my main concerns with this manuscript, and I believe it is ready for publication after some language edits.

Upon reading the response to reviewers, referee #2's comments, and thinking some more, I now agree that this data product will be of wide use. Even if many people do use GLODAPv2 instead of this data product, this paper still makes an important comment about traceability. Moreover, the added comparisons to GLODAPv2 will help the readers put into context how different this data product is and understand how much GLODAPv2 has to gain through a deeper consideration of traceability.

A: Many thanks for positive evaluation to my revised draft especially in terms of traceability. I also thank reviewer#1 for his/her time to read and check my draft very carefully and make suggestions to improve my draft. I believe that I amended my draft based on your suggestions. I also add a few sentences to make clear this article regarding with your comments.

Reply to specific suggestions and confusions:

P1L15: "used as a reference"

A: I did so.

P1L16: "...by other cruises SI traceable." "data was an additional"

A: I amended as below including next comment.

Cruises that used certified reference materials or reference materials (CRMs/RMs) for seawater nutrient concentration measurements were used as a reference of unbroken chain of comparison to determine correction factors which made nutrient concentrations obtained by other cruises SI traceable. Dissolved oxygen was secondarily-quality controlled using the same methodology as was used to create the nutrients gridded data product, but, lacking a traceable standard, the resulting oxygen data product is not SI traceable.

P1L17: "Dissolved oxygen was secondarily-quality controlled using the same methodology as was used to create the nutrients gridded data product, but, lacking a traceable standard, the resulting oxygen data product is not SI traceable."

A: I amended as below including previous comment.

Cruises that used certified reference materials or reference materials (CRMs/RMs) for seawater nutrient concentration measurements were used as a reference of unbroken chain of comparison to determine correction factors which made nutrient concentrations obtained by

other cruises SI traceable. Dissolved oxygen was secondarily-quality controlled using the same methodology as was used to create the nutrients gridded data product, but, lacking a traceable standard, the resulting oxygen data product is not SI traceable.

P1L19: “as a SI”

A: I did so.

P2L18: “after a CRM for nutrients”

A: I did so.

P3L5: Consider putting the 2nd paragraph at the end of the first.

Yes, I did so.

P3L10: “correction factors for silicate”

A: I did so.

P4L6: “oxygen concentrations”

A: I did so.

P4L7: “world have some”

A: I did so.

P4L9 “and an unbroken”

A: I did so.

P4L16: “data, specifically the unbroken chain of comparison, which”

A: I did so.

P4L17: “cruises identified in section 2. As noted, this does not mean the oxygen product is SI traceable.”

A: I did so.

P5L6: “an accurate baseline of”

A: I did so.

P5L7: “set to 1.00, indicating no adjustment is applied, because...”

A: I did so.

P5L9: "aimed at being consistent with the data obtained"

A: I did so.

P5L17: "through an inter-laboratory" or through inter-laboratory comparison studies"

A: I did so.

P5L18: "2010), these two"

A: I did so.

P6L12: "cover the regions not covered by category 1 and 2 cruises, categories" Southern Hemisphere should be capitalized.

A: I did so.

P6L16: "3 times the standard" ... were these calculated for each cruise? If so, indicate.

A: Exact explanation on this issue is as follows.

3 times the standard deviation was calculated in a selected region of which size was 5 degree of latitude and 5 degree of longitude, and of which layer thickness was 2 times to 6 times of layer thickness at corresponding depth of 136 layers used in this study as standard depths of data product, eg. 10, 20, 30, 40, 50, 75, 100, 125, 150,175, 200, 250, 300,,,, 6450 and 6500 meters. At higher latitude, longitude disntance expanded and it kept 500 km width. This size is consistent with a 250-km radius of crossover points. As an example of region setting, latitude between 20 deg. N and 25 deg. N and longitude between 140 deg. E and 148 deg. E and depth between 950 meter and 1150 meter were set for a region of median filter with a criteria of 3 times the standard deviation of the nutrients data in the region.

Therefore, I add a sentence in the main text as below.

This median filter procedure was done to data sets after the nutrients data was divided into sets of data which was involved in a region of which size was about 500 km x 500 km square and of which thickness was 100 meter to 300 meters in the deep ocean and 10-50 meters in the shallow ocean.

P6L17: "data. Questionable"

A: I did so.

P6L18: "and creation of the global gridded data product."

A: I did so.

P6L21: “243 cruise crossover points”

A: I did so.

P6L22 through P7L2: I don’t understand this text and it sounds important.

The original sentence was not correct. The author wanted to state a few exceptions about crossovers constructed only category 3–7 cruises in the in the Pacific sector of the Southern Ocean. This issue is already stated in Page 6 Line 12 as “In southern hemisphere, to cover the some sea areas where categories 1 and 2 were not there, categories 3-7 were used.” Therefore, I removed the sentence in Page 6 Line 12 and revised the sentence here as below to make clear the explanation of exception on crossovers.

In the Pacific sector of the Southern Ocean, to cover the regions not covered by category 1 and 2 cruises, categories 3-7 were used to construct crossovers.

P7L2: “As an example, Figure 5 shows the locations of the subset of P03 and P14 stations used for a crossover comparison for a crossover at···(Include East/West for longitude). This crossover is assigned the designation CR081E, and there were two category···”

A: I did so.

P7L10: “on the 30”

A: I did so.

P8L5: Words are missing here··· maybe “because there was a smaller”

A: Yes, I add “there was”.

P8L10: “nutrient concentrations”

A: I did so.

P8L12: “measurements with a 250-km radius becomes very small. When in-house standards were used, station-··· may contribute meaningfully”

A: I did so.

P8L15: CV is defined twice. It should probably be “the CV” in most places.

A: I have deleted second one and amended as “the CV” in all places.

P8L20: “concentrations was similar”

A: I did so.

P9L4: “oxygen and comparability was kept only with potassium iodate solutions.”

A: I did so.

P9L6: “nutrient concentrations”

A: I did so.

P9L8: “category 1 cruises, as”

A: I did so.

P9L10: “250-km radius (based on the data···crossover points)”

A: I made correction following all suggestions stated above for P8L20 to P9L10..

P9L15: “uncertainty from both measurement”

A: I did not follow this suggestion and revised the sentence as below to make clear the discussion.

The larger mean of the standard deviations of the integrated values for the four parameters at crossovers for the cruises in categories 2–7 might reflect the relatively larger uncertainty which was combined of uncertainty of measurement and within-cruise variability (= variability of measurements among several stations within a 250-km radius).

P9L17: “When we apply the method adopted in this study, we need to consider uncertainties of measurements”

A: I did so.

P10L3: What is meant by “with uncertainty” ?

A: This uncertainty is of correction factor, so I revised this sentence as below.

Based on the wider coverage by the cruises in category 2, those cruises were used as secondary key cruises after correction factors and their uncertainties were applied to the integrated values.

P10L19: “of 50”

A: I did so.

P11Step1: “First, nutrient and oxygen profiles were interpolated vertically to 136 levels.”

I think we need original first step and also make clear what I did, then I revised this part as

below.

Step 1: Profiles of which factor were determined were selected to create the global gridded dataset. Then nutrients and oxygen concentrations were corrected by the factors.

Step 2: Nutrient and oxygen profiles were interpolated vertically to 136 levels.

And original Step2 put Step3

P11L8: “deg. E. Then the gridded data surface function of the GMT was used to map these interpolated data horizontally for each of the 136 layers.” These two steps both need to be rephrased.

A: I did so.

Table 4: "Dissolved oxygen

A: I had corrected this typo-mistake.

Figure 5: What are the other squares at the top of the figure? What are the Lat/Lon units (degrees N/S, E/W)

A: I deleted several squares those should not be in the figure.

I amended labels of X- and Y- as you mentioned.

Figure 6: use ½ as many axis labels for the lower left panel.

Honest saying, I cannot understand exact meaning of this comment, but I guess that I need to delete “13.5, 14.5,,,” to reduce number of labels of x-axis and I did so.

End of reply.

Global CRM/RM-scaled nutrient gridded dataset GND13

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Abstract. A global nutrients gridded dataset that might be the basis for studies of more accurate spatial distributions of

10 nutrients in the global ocean was created and named GND13. During 30 cruises, reference materials of nutrients in seawater

or their equivalents were used at all stations, and high-precision measurements were made. The precision of the nutrient

analyses was better than 0.2%. Data were collected from the hydrographic cruises in JASMTEC R/V *Mirai* cruises, JMA

cruise, CARINA, PACIFICA and WGHC datasets from which nutrient data were available. Analyses were conducted at 243

crossover stations. Cruises that used certified reference materials or reference materials (CRMs/RMs) for seawater nutrient

15 concentration measurements were used as a reference of unbroken chain of comparison to determine correction factors

which made nutrient concentrations obtained by other cruises SI traceable. Dissolved oxygen was secondarily-quality

controlled using the same methodology as was used to create the nutrients gridded data product, but, lacking a traceable

standard, the resulting oxygen data product is not SI traceable. Finally, a dataset of nitrate, phosphate, silicate concentrations

was created at latitude and longitude intervals of 0.5° and on 136 isobaric surfaces to depths of 6500 meters as a SI traceable

20 dataset. This dataset has already been published at doi:10.17596/0000001.

1 Introduction

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削除: Dissolved oxygen concentration data was additional parameter of GND13 using same methodology to create nutrients gridded data, but not traceable to SI.

Global oceanic biogeochemical cycles are being significantly altered by the direct and indirect impacts of human activities. It is therefore necessary to obtain accurate information about changes and trends of concentrations of inorganic carbon and dissolved inorganic nutrients in both shallow and deep ocean waters. For this information to be of practical use, it is critical that results from different laboratories can be compared with complete confidence. A global consensus about nutrient concentrations requires that there be access to certified reference materials (CRMs), and there must be a requirement or ethos for the use of these CRMs when oceanic nutrient concentrations are measured and subsequently when they are recorded in global databases, incorporated in climate models, and ultimately used to quantify changes to the Earth system.

The 2007 IPCC Report highlighted the problem inherent in comparing datasets by stating that “Uncertainties in deep ocean nutrient observations may be responsible for the lack of coherence in the nutrient changes. Sources of inaccuracy include the limited number of observations and the lack of compatibility between measurements from different laboratories at different times” (Bindoff et al., 2007). Analyses of nutrient concentrations from crossover stations have shown consistent disagreement of up to 10 % for deep water nutrient data during the last three decades (Aoyama et al., 2013; Tanhua et al., 2009). Results of inter-laboratory comparison studies since 2003 have shown biases of a similar magnitude between some participant laboratories (Aoyama et al., 2007; 2008; 2010; 2016; 2018). This pattern indicates that analytical problems may be the main cause of the large discrepancies in reported deep water nutrient concentrations. The reported results imply that these biases are also present throughout the water column. These comparisons were based on only a small number of specific studies, but there are many oceanic nutrient datasets reported, published, and stored on international databases with no references to CRMs at all. Although this situation has improved somewhat since 2011 after a CRM of nutrients became available, it is still difficult to ascertain with total confidence any temporal changes in oceanic nutrient concentrations. We can now detect changes in deep ocean temperature (and hence heat content) (Levitus et al., 2009; 2012; Kouketsu et al., 2009; Rhein et al., 2013) because of the excellent comparability of temperature measurements over a number of years. Changes to the carbonate system parameters in the deep ocean have also been reported with comparability ensured by the use of CRMs (e.g., Wanninkhof et al., 2010). Similarly, changes in oceanic oxygen concentrations can now be determined (Stendardo and Gruber, 2012).

Reference materials (RMs) and CRMs for nutrients in seawater have been developed for oceanographic use. These currently include a Danish RM (Eurofins), NRC-Canada CRM (MOOS-3), a new RM developed by Korea (K-RMS), and one developed by KANSO-Japan. The reference material for nutrients in seawater produced by KANSO has been used in the inter-laboratory comparison exercise organized by Meteorological Research Institute, MRI, and jointly by International Ocean Carbon Coordination Project, IOCCP and Japan Agency for Marine-Earth Science and Technology, JAMSTEC, since 2003 (Aoyama et al., 2007; 2008; 2010; 2016; 2018). The results of the latest inter-laboratory comparison exercise (IC),

“IOCCP-JAMSTEC 2017/18 Inter-laboratory Calibration Exercise of a Certified Reference Material for Nutrients in Seawater”, are now available (Aoyama et al., 2018). It is clear from the current results (see Figs. 6, 7, and 8 in the 2017/2018 report) that the normalized cumulative distributions of nitrate and phosphate were better in 2018 than in previous years. The curves were flatter than the normalized cumulative distributions in previous IC exercises. This improvement for nitrate and phosphate measurements might be a reflection of the fact that the number of laboratories that use CRM/RMs was increasing during those years.

移動 (挿入) [1]

The implication is that comparability of silicate analyses among the laboratories did not improve between 2008 to 2018 to the same degree that it did for nitrate/phosphate, and the correction factors for silicate were indeed more variable and uncertain than the correction factors for nitrate and phosphate.

上へ移動 [1]: This improvement might be a reflection of the fact that the number of laboratories that use CRM/RMs was increasing during those years.

This difference of comparability between nitrate/phosphate and silicate analyses can be also seen in the results of correction factors estimation with uncertainty in this study. In particular, correction factors of silicate were more variable and were associated with greater uncertainty than the correction factors for nitrate and phosphate. Consensus standard deviations of nutrient concentrations of nitrate, phosphate and silicate were one order of magnitude larger than the homogeneity of the currently available CRM/RMs and were about double the reported precision of measurements of the individual laboratories.

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These IC results therefore showed that use of CRMs should greatly improve the comparability of nutrient data among laboratories throughout the world. The current high level of analytical performance at many participating laboratories indicates that the use of certified reference materials would establish traceability. The use of CRM/RMs during global cruises in the CLIVAR (Climate and Ocean: Variability, Predictability and Change), GO-SHIP (Global Ocean Ship-based Hydrographic Investigations Program), and GEOTRACES projects has been increasing, and the author has been using CRM/RMs during the cruises of the Japan Agency for Marine-Earth Science and Technology (JAMSTEC) R/V *Mirai* since 2003. Disagreements between cruises at depth tend to be smaller when reference materials are used.

On the other hand, the method for determining the dissolved oxygen concentration in seawater is generally the Carpenter method (Carpenter, 1965), which is an improvement of the Winkler method, but is hereafter simply referred to as the Winkler method. In this Winkler method, manganese hydroxide “fixes” dissolved oxygen under alkaline conditions, and

the “fixed” dissolved oxygen quantitatively oxidizes iodine ions to free iodine under acidic conditions. Titrating the free iodine with a sodium thiosulfate solution of known concentration indirectly quantifies the dissolved oxygen concentration.

The sodium thiosulfate solution concentration is determined by titration of a potassium iodate solution of known concentration (potassium iodate quantitatively oxidizes iodine ions to free iodine under acidic conditions). In Japan, SI-traceable certified reference potassium iodate standards are supplied by the National Meteorology Institute of Japan, National Institute of Advanced Industrial Science and Technology (NMIJ). Ocean Scientific International Ltd, OSIL, UK, and FUJIFILM Wako Pure Chemical Corporation, Japan, also provides Potassium Iodate solutions, which are used to standardize the thiosulfate solution in the widely used Winkler titration method. Therefore, dissolved oxygen concentrations measured around the world have some extent of comparability.

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This article describes a global gridded dataset produced using CRM/RM-scaled SI traceable nutrient concentrations based on key cruises that used CRM/RM and an unbroken chain of comparison. Several previous publications have provided synthesis results of data collected by several projects such as the Global Ocean Data Analysis Project (GLODAP, GLODAPv2, GLODAP v2 update), Carbon in Atlantic Ocean (CARINA), and Pacific Ocean Interior Carbon (PACIFICA) projects (Key et al., 2009; Suzuki et al., 2013; Olsen et al., 2016; Olsen et al., 2019). The time frame of this work is that cruises categorized 1 were conducted between 2003 and 2013, all of the resulted data in this work are adjusted for 2003-2013 time frame. Another positive attribute of this work is that uncertainty of correction factors could be estimated.

The author also adds dissolved oxygen concentration data as additional parameter of GND13 using same technology to create nutrients gridded data, specifically the unbroken chain of comparison, which means obtained gridded data of dissolved

削除: data, unbroken chain of comparison, which

oxygen are traceable to a set of data obtained from 30 key cruises identified in section 2. As noted, this does not mean the oxygen product is SI traceable.

削除: cruises stated in chapter 2 and did not mean SI traceable.

This article is an effort to establish a global nutrients dataset for which comparability and traceability in space and time are explicitly ensured based on the use of CRM/RMs of nutrients in seawater. Another positive attribute of this work is that uncertainty of correction factors could be estimated.

2 Methods and Data

Data from 30 cruises that used CRM/RM for quality control of nutrient concentrations in seawater were used to obtain an accurate baseline of the spatial distribution of nutrient concentrations in the ocean. The correction factors for those cruises were set to 1.00, indicating no adjustment is applied, because comparability of nutrient concentrations was ensured (Sato et al., 2010).

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削除: set to 1.00 because

For oxygen data, the factors for 30 cruises were assumed to be 1.00 because gridded data of dissolved oxygen are aimed at being consistent with the data obtained from 30 key cruises.

削除: aimed to be traceable to a set of data obtained

2.1 Data collection and quality control

Nutrient data from the global ocean were collected from various sources and separated into categories from 1 to 7 (Table 1). Thirty cruises were assigned to category 1. Twenty-five of the 30 key cruises were carried out by R/V *Mirai* during 2003–2013. The author used RM/CRM on those cruises as working standards for nutrient measurements at all stations to ensure high quality and comparability among the stations and among the cruises. In the Atlantic Ocean, five cruises were also selected as category 1 because RM were used on two of the five cruises. Since comparability of nutrients data between JAMSTEC R/V *Mirai* cruises during the period from 2003 to 2013 and NIOZ cruises conducted in 2005 and 2007 was

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explicitly confirmed through inter-laboratory comparison studies for reference materials of nutrients in seawater conducted in 2006 and 2008 (Aoyama et al., 2008; 2010), these two cruises were also added to category 1 to increase coverage by

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category 1 cruises in the Atlantic Ocean. Most of the data in category 2 were obtained from the CARINA project dataset.

Most of the data in category 3 were obtained from the PACIFICA project dataset for the period 1991–2008. Many data were

obtained from the World Ocean Circulation Experiment (WOCE) Global Hydrographic Climatology (WGHC) (Gouretski

and Koltermann, 2004) data product. That dataset includes data from many cruises during the period 1925–1996, and those

data were assigned to category 4. Category 5 was intentionally blank for future use. Some cruises conducted by the Japan

Meteorological Agency (JMA) were not included in the CARINA, PACIFICA and WGHC datasets. They were therefore

included in the dataset for this study and assigned to category 6. Data from about 80 cruises by the JMA and United States

institutes that were not included in the above categories were assigned to category 7.

Figures 1–4 show the locations of all the stations where data were collected. In these figures, stations in category 1 are marked in dark blue, stations in category 2 are marked in light blue, and stations in categories 3–7 are marked in red. It is

apparent from these figures that the category 1 cruises did not cover the whole ocean, but if category 1 and 2 data are used to

create a global dataset, the spatial coverage increases to almost all of the ocean, and the resultant dataset is a high quality

global nutrient dataset.

削除: In southern hemisphere, to cover the some sea areas where categories 1 and 2 were not there, categories 3-7 were used.

It is important to do quality control before using this historical dataset because it contains questionable data. In the WOCE dataset and later, there are quality flags (WOCE Hydrographic Programme Office, 1994). Only data associated with quality flag 2 (i.e., data quality is good) were therefore used in this study. Because the historical data and some of the data did not have quality flags, a median filter of which criteria is 3 time of standard deviation was criteria for outliers was used to

identify questionable data. This median filter procedure was done to data sets after the nutrients data was divided into sets of data which was involved in a region of which size was about 500 km x 500 km square and of which thickness was 100 meter to 300 meters in the deep ocean and 10-50 meters in the shallow ocean. Questionable data were removed from the dataset

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before vertical integration, estimation of correction factors and creation of the global gridded data product.

削除: and create global gridded data.

2.2 Crossover analysis

In general, stations from each cruise within 250 km of 243 points worldwide were selected if there were data from several stations from at least a cruise in category 1 and at least, respectively a cruise from category 2. In the Pacific sector of the Southern Ocean, to cover the regions not covered by category 1 and 2 cruises, categories 3-7 were used to construct crossovers. As an example, Figure 5 shows the locations of the subset of P03 and P14 stations used for a crossover comparison for a crossover at 24.2°N and 179°E. This crossover is assigned the designation CR081E, and there were two category 1 cruises and two category 3-7 cruises. Supplementary Fig. S1 shows all station locations at the 243 crossovers.

Figure 6 shows examples of vertical profiles of nitrate concentrations, phosphate concentrations, nitrate-to-phosphate concentration ratios, and silicate concentrations at crossover CR081E. Figure 6 also shows climatological nutrient concentrations in the WGHC dataset and WOA05 dataset for comparison. There were two cruises conducted in 2005 and 2007, which were category 1 cruises, where RMs were used as working standards at all stations by the author. The results from those cruises were in good agreement with data collected within a 250-km radius, and the error bounds (i.e., uncertainties) overlapped completely. In contrast, concentrations from two cruises conducted in 1985 and 1993 were relatively scattered (Fig. 6). To estimate correction factors based on the 30 key cruises, vertical integration between depths of 1000 meters and 2000 meters, 1500 meters and 2500 meters and 2000 meters and 3000 meters for nitrate, phosphate, silicate and oxygen were done at all stations within each of the 243 crossovers. This integration was done based on the Akima interpolation method (Akima, 1970). When the number of profiles of a cruise exceeded 3, the standard deviation of the integrated values was calculated as a metric of uncertainty of correction factor. Table 2 shows the valid number of profiles obtained by vertical integration from 1000 meters to 2000 meter depths, from 1500 meter to 2500 meter depths from 2000 meters to 3000 meter depths for nitrate, phosphate, silicate, and oxygen concentrations at the P03-P14 crossover stations at 24.2°N and 179°E. As expected from the vertical profiles at the crossovers, the integrated values in units of $\mu\text{mol m}^{-2}$ for the

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削除: A few exceptions were crossovers in the Pacific sector of the Southern Ocean, where crossovers were selected from category 2 cruises and category 3-7 cruises or among category 3-7 cruises to expand coverage.

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削除: Figure 5 shows an example of station locations at P03-P14 crossovers at

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two cruises in category 1 (e.g., 49MR0505_2_1 and 49MR0706_1_) agreed to within two standard deviations for all four parameters. The standard deviations of two category 2 cruises in 1985 and 1993 were relatively large in general, and there were systematic differences that have already been identified in previous synthesis work. Because there was assurance of comparability of nutrient concentrations among the 30 key cruises, the author set the correction factor for these cruises to 1.00. Because the measurement uncertainties during these cruises were less than 0.5% in general, the uncertainty of correction factors were assumed to be 0.00.

To estimate correction factors at all crossovers the author selected to use integrated values by vertical integration from 1500 meters to 2500 meter depths because there was a smaller Coefficient of Variation (CV), a ratio of the standard deviations of the integrated values to a mean of the integrated value and largest total number of the CV among three integration ranges for nitrate, phosphate and silicate.

The standard deviation of the integrated values for a set of profiles from each cruise within crossovers can be considered as the combined uncertainty of measurement uncertainty at each profile, station-station variability of measurement within a 250-km radius and natural variability of nutrient concentrations among several stations within a 250-km radius at crossovers.

It is expected that when RM/CRM were used as working standards to get a calibration curve, station-station variability of measurements with a 250-km radius becomes very small. When in-house standards were used, station-station variability of measurement within a 250-km radius may contribute meaningfully. Therefore, it is interesting to look at the CV of the four parameters (Table 3). Figure 7 also shows histograms of the CV of integrated value of the nitrate data in categories 1–7. It is very clear that the mean of the CV of integrated values were 0.005 for nitrate and phosphate for category 1 cruises and that for silicate was 0.009. The means of the CV of integrated values for nitrate, phosphate and silicate were smaller than those for categories 2–7. The main cause of the smaller mean of the CV of the integrated values for nutrient concentrations

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删除: measurement within a 250-km radius becomes very small while in-house standard was used, station-

删除: may contribute to increase combined uncertainty

删除: Coefficient of Variation (CV), a ratio of the standard deviations of the integrated values to a mean of the integrated value

measured during the category 1 cruises might be the use of CRM/RM. The mean of the CV of the integrated values for nutrient concentrations was, similar to the precision of each measurement, roughly 0.2–1.0%. It should be also noted that the silicate measurements were compromised by some difficulties and/or instabilities—unlike the nitrate/phosphate measurements—that were observed in the global IC study discussed in the introduction of this article. On the other hand, the corresponding values for category 1 oxygen measurements were similar to those for category 2–7 cruises because there are no seawater matrix reference materials for dissolved oxygen and comparability was kept only with potassium iodate solutions.

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削除: oxygen exist and comparability was kept by potassium iodate solution worldwide as similar magnitude.

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During the timeframe of this study from 2003 to 2013, temporal variation of nutrient concentrations within a 250-km radius at crossovers at 1500-2500 meter depth was very small and it could be assumed to be negligible based on comparison at crossovers between/among category 1 cruises, as shown in Figure S1 especially in the Pacific Ocean. Natural variabilities of nutrients within a 250-km radius at 1500-2500 meter depth were similar to or smaller than the combined uncertainty of uncertainty of measurement and station-station variability of measurement within a 250-km radius based on the data in Table 3 and other crossover points. In other words, deep sea water within a 250-km radius at 1500-2500 meters was quite homogeneous horizontally, and the variability of nutrient concentrations observed in category 2 and 4 cruises might be due to the lower comparability of the nutrient measurements made during those cruises. The larger mean of the standard deviations of the integrated values for the four parameters at crossovers for the cruises in categories 2–7 might reflect the relatively larger uncertainty which was combined of uncertainty of measurement and within-cruise variability (= variability of measurements among several stations within a 250-km radius).

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When we apply the method adopted in this study, we need to consider uncertainties of measurements and within-cruise variability (= variability of measurements among several stations within a 250-km radius) that might cause the correction

factors to be uncertain. The uncertainties of the correction factors were estimated in terms of the CV of the integrated values within crossovers as a first step in estimation of correction factors. Key cruises included two-thirds of the 243 crossover points (Figs. 1–4). To estimate correction factors at the remaining crossover points, correction factor estimations were done progressively. Based on the wider coverage by the cruises in category 2, those cruises were used as secondary key cruises after correction factors and their uncertainties were applied to the integrated values. Factors for cruises in categories 3–7 were then estimated, with the exception of several crossover stations. Supplementary Table 1 shows estimated factors and their uncertainties for all cruises.

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Comparisons were made between the factors obtained in this study and in GLODAP v2 (Olsen et al., 2016;2019).

Figures 8–11 show the results. For nitrate and phosphate, the correction factors obtained in this study were in good

agreement with those obtained by GLODAP v2 when the correction factors relatively deviated far from 1.00. The

implication is that both synthesis work and direct comparisons as done by this study can detect differences between cruises

and estimated correction factors correctly when the nutrient concentrations obviously differ from values obtained on nearby

cruises. For many GLODAP v2 cruises, factors were assigned a value of 1.00, but it is obvious that direct comparisons

resulted in factors that were slightly larger or smaller (Figs. 7–9) because synthesis work could not identify differences

among cruises if those differences were not large. Direct comparisons, however, could determine correction factors with

uncertainties more precisely. In general, the differences of the correction factors obtained by two methods, synthesis like

GLODAP v2 and direct comparison as this study, for nitrate and phosphate were around +0.02 and –0.04, whereas the

differences for silicate were relatively large: ± 0.06 . For oxygen, the differences were much larger: ± 0.10 .

2.3 Gridded dataset

Based on the factors obtained in this study, a dataset was created at latitude and longitude intervals of 0.5° and on 136 isobaric surfaces at intervals of 50 meters. The uncertainties of the nutrient concentrations were about 2% for nitrate and phosphate and 5% for silicate and oxygen. This uncertainty was equated to twice the standard deviations of the integrated values for the category 2 cruises. The following steps were used to create the global gridded dataset.

Step 1: Profiles of which factor were determined were selected to create the global gridded dataset. Then nutrients and oxygen concentrations were corrected by the factors.

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Step 2: Nutrient and oxygen profiles were interpolated vertically to 136 levels.

删除: and vertical interpolations were then

删除: done for each profile on 136 layers.

Step 3: To have smooth gridded data at 0 deg. E (=360 deg. E), data obtained step 1 for 0 deg. E to 20 deg. E were copied to 360 deg. E to 380 deg. E region and data for 340 deg. E to 360 deg. E were copied to -20 deg. E to 0

删除: 2

删除: o

deg. E. Then a surface function of The Generic Mapping Tools, GMT (<https://www.soest.hawaii.edu/gmt/>),

删除: o create griddeddata

GMT was used to map these interpolated data horizontally for each of the 136 layers. North of 65°N, the

删除: were carried out on each of the 136 layers .

latitude and longitude of the data points were converted to an X–Y surface. Then conduct a surface function of GMT for each depth. Convert the gridded data in the X–Y plane to latitude and longitude at 0.5° intervals.

A gridded dataset with 136 layers at latitude and longitude intervals of 0.5°—the Global Nutrient Dataset 2013— was

then created. Figure 12a–d shows the horizontal distributions of nitrate, phosphate, silicate, and oxygen concentrations at a depth of 1800 meters in the Pacific Ocean as an example.

To determine the total amount of nitrate, phosphate, silicate, and oxygen in the ocean, the volume and area corresponding to each grid point were calculated using the ETOPO 2 topographic, bathymetric dataset (2-minute mesh). The concentrations were multiplied by the volume of the obtained grid point to find the number of moles of nitrate, phosphate,

silicate, or oxygen at each grid point, and the results were summed for each sea area. In this way the total amounts of nitrate, phosphate, silicate, and oxygen in the ocean were estimated as well as the associated uncertainties.

3 Results

This dataset was designated the Global Nutrients Dataset 2013 (GND13). The GND13 is already available at doi:10.17596/0000001 on the JAMSTEC web site. The Fortran source code and ctl script of Grads are also available at the site. All figures of the horizontal distributions of nitrate, phosphate, silicate, and oxygen are available as supplementary figures of this article.

Table 4 shows the total mass in petagrams of nitrate, phosphate, silicate, and dissolved oxygen in the ocean. Using the same methodology, the total petagrams of nitrate, phosphate, silicate, and dissolved oxygen were also calculated for the WOA09 (Garcia et al., 2010a;2010b) and WGHC datasets. The total amounts of nitrate, phosphate, silicate, and dissolved oxygen \pm uncertainty were 573 ± 11 Pg N, 89.0 ± 1.8 Pg P, 3300 ± 170 Pg Si and 7180 ± 360 Pg O₂, respectively. As can be seen in Table 4, the results of GND13 were consistent within uncertainty to the total amounts calculated from the WOA 09 and WGHC climatological concentrations, which had been published previously and were the initial values of various studies based on a current ocean general circulation model. The total amount of nitrate by GND13 was large compared with the literature values: 541 Pg N by Sarmiento and Gruber(2006) and close to 570 Pg N by Wada and Hattori (1990). The medians of the N:P molar ratios at depths >2 km were 14.6 for WOA 09 and 14.3 for GND13, and in the latter case the distribution shows high kurtosis (figure not shown). The implication is that the previous lattice point dataset was generated from a dataset with less comparability, whereas the GND13 dataset was generated from a dataset with higher comparability.

4 Data availability

The GND13 is available at doi:10.17596/0000001 at the JAMSTEC web site

http://www.godac.jamstec.go.jp/catalog/data_catalog/metadataDisp/GND13?lang=en.

5 Conclusions

A global nutrients gridded dataset was created and named GND13. Thirty cruises incorporating reference materials for nutrients in seawater or their equivalent were used. The precision of the nutrient analyses was better than 0.2%, and comparability between stations was ensured. Nutrient data were collected from all of the hydrographic cruises from which nutrient data were available. Crossover analyses were conducted at 243 crossovers where data from our cruises served as references to determine factors for adjusting nutrient concentrations obtained during other cruises. Dissolved oxygen concentrations were included as an additional parameter in the dataset using the same protocol. Finally, global datasets of nitrate, phosphate, silicate, and dissolved oxygen concentrations were created at 0.5° latitude and longitude grid points on 136 isobathymetric layers to a depth of 6.5 km. This dataset will facilitate studies of the behavior of carbon:nitrogen:phosphorus:oxygen stoichiometry in the ocean in the near future.

6 Appendices

Supplementary figures

Supplementary tables

7 Supplement link (will be included by Copernicus)

xxx

Author contribution

Michio Aoyama is the only scientist who created the dataset GND13.

Competing interests

The author declares that he has no conflict of interest.

Disclaimer

xxx

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