Comparisons of Summer-Winter Carbonate and Nutrient Data in the Southwest Indian Ocean

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ABSTRACT

Chemical data were obtained in the southwestern Indian Ocean in the austral Winter (July) of 1984 and in the austral summer (Feb. - March) of 1985 aboard the French research vessel, MARION DUFRESNE. The winter data represent initial chemical concentrations in an important source region of the Antarctic Intermediate Water at the time of its formation. For the first time, one can evaluate variations in the chemical cycles in the Antarctic Intermediate Water of the Indian Ocean with reference to the source water in winter. These winter data were compared with our summer data and with the summer data obtained in 1978 during the GEOSECS Expedition. Preliminary results indicate the following: the surface pH and normalized nitrate, alkalinity and total CO_2 values are found to correlate linearly with temperature; small deviations from the linearity are related to the Subantarctic and Subtropical Fronts and to the equatorial upwelling; large variations in nitrate and pH are found in surface waters collected at the same location but in different seasons; however, there is less variation between pH or normalized nitrate concentrations when compared at the same temperature; a seasonal difference in alkalinity and total CO_2 may exist, even when compared at the same salinity and temperature; the decrease in alkalinity and total CO_2 between the Antarctic Waters and the Indian Central Water found north of the Subtropical Front can perhaps be attributed to the decrease in nitrate and the increase in temperature; the remnant North Atlantic Deep Water (NADW), which has a very weak salinity signal, is identified clearly by pH and total CO_2 data; and nutrient and oxygen data also help in tracing NADW.

1. INTRODUCTION

Deep waters from the three oceans move to the Southern Ocean and mix there. The resultant, relatively homogeneous water becomes the major source of the Antarctic Bottom Water (AABW), which spreads back out into the deep world oceans. Consequently, the chemistry of Southern Ocean water is a baseline for the deep world oceans. One must therefore know the chemistry of the Southern Ocean water in order to understand global biogeochemical cycles (Wust, 1939; Bolin, 1983; Chen, 1984). Unfortunately, only a few high-precision chemical oceanographic programs have been conducted in the Indian Ocean

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section of the Southern Ocean. For instance, I know of only 7 stations with good quality carbonate and nutrient data south of 30° S prior to our investigations (Weiss *et al.*, 1983). Not knowing the characteristic properties of the water near its origin, therefore, makes it difficult, if not impossible, to interpret variations in the carbon chemistry or to calculate excess CO_2 in the Indian Ocean. Furthermore, the scant data in the southern Indian Ocean were all collected in the summer; whether the summer data are representative of the mainly winterformed waters is entirely uncertain (Chen and Pytkowicz, 1979; Chen, 1982a,b; 1987a,b; 1988b; Chen and Poisson, 1984; Poisson and Chen, 1987).

Recognizing the importance of collecting more chemical data, especially seasonal data, in the southern Indian Ocean, I participated in two cruises, INDIVAT 1 and INDIGO 1 / INDIVAT 3. INDIVAT (INDIEN VALORISATION de TRANSIT) took advantage of the travel schedule of the French research/supply vessel, MARION DUFRESNE, which steams from la Reunion to Crozet, Kerguelen, Amsterdam, and back to la Reunion three to four times per year. The French TAAF (TERRES AUSTRALES et ANTARCTIQUES FRANCAISES), which operates the vessel, agreed that I could use the ship to collect and measure surface samples during transit. Because the vessel crosses the Subtropical and Antarctic Fronts many times a year, it provides an excellent opportunity to study the seasonal variation of chemicals in the formation region of the Antarctic Intermediate Water (Chen and Poisson, 1986; Chen *et al.*, 1986).

TAAF also agreed to permit me to reoccupy two GEOSECS stations (GS 427 and 429) during each INDIVAT cruise. Deep samples provide me with a means to calibrate my results for comparison with data reported in the literature. In addition, I can determine how much the seasonal variation affects the water column. Such information is essential for comparisons from year to year.

The second program, the INDIGO (INDIEN GAZ OCEAN), involves the use of the MARION DUFRESNE to collect deep samples in different regions of the Indian Ocean once a year for a minimum of four years.

The first INDIVAT expedition took place in July 1984. INDIVAT 2, however, had to be cancelled owing to logistical problems. The combined INDIGO 1 / INDIVAT 3 expedition was carried out in February/March 1985.

2. OUTLINE OF THE EXPEDITIONS

The MARION DUFRESNE departed from la Reunion for the INDIVAT 1 Expedition on 3 July 1984, reoccupied GS 427 on 5 July, and reoccupied GS 429 on 19 July after a stop in Crozet. The vessel then proceeded to Kerguelen and Amsterdam Is. and returned to la Reunion on 4 August. The cruise track is shown in Figure 1. The Subtropical Front was near 40° S and the Antarctic Front was near 47° S. While underway, sea surface samples were collected hourly from a seawater intake located at the bow 4 *m* below the surface. Temperature and salinity at the intake were recorded by a Thermosalinograph. During INDIVAT 1 seawater was pumped through a rubber tube to a van near the laboratory where samples were taken. Because of the delay in sampling, the recorded temperature and salinity may not



Fig. 1. The cruise tracks of INDIVAT 1 (dashed line) and INDIGO 1 / INDIVAT 3 (solid line).

always match the samples taken, especially near an oceanic front. The long time (~5 *minutes*) required to collect all samples (salinity, oxygen, *pH*, alkalinity, total CO_2 , pCO_2 , nitrate, phosphate, and silicate) may also have caused some discrepancies in the data, as the water flowing out of the tube at the beginning of sampling may have been somewhat different from the water flowing out at the end of sampling. Subsurface samples at GS 427 and 429 were obtained using a Neil Brown CTD-Rosette system (Chen *et al.*, 1986).

The first INDIGO expedition (with emphasis on subsurface samples) and the third INDIVAT expedition (with emphasis on underway surface samples) were combined. R/V MARION DUFRESNE departed from la Reunion on 23 February 1985 and returned on 30

March. Chemical data were collected from 23 stations including four GEOSECS stations (427 – 429, 454). Underway samples were also taken directly near the underwater intake. The attempt to reach the Antarctic Continent, however, was thwarted by foul weather. The cruise track is given in Figure 1. The Subtropical Front was at approximately 43° S and the Antarctic Front was near 52° S (Chen *et al.*, 1986).

A large number of physical and chemical properties were measured, including temperature, salinity, oxygen, nitrate, phosphate, silicate, pH, alkalinity, total CO_2 , pCO_2 , calcium, magnesium, boron, Kr-85, tritium, C-14, freons, and particulates (Chen and Poisson, 1986; Chen *et al.*, 1986). I only report the carbonate and nutrient data here because other data will be discussed elsewhere (e.g. Goyet *et al.*, 1990).

3. EXPERIMENTAL TECHNIQUE

The *pH* samples were all analyzed at $25 \pm 0.02^{\circ}$ C with a Radiometer combination electrode within 30 *minutes*. NBS 4.004 and 7.415 buffers were used to calibrate the electrode. The reproducibility of the *pH* measurements is better than ± 0.003 units for replicate samples. The electrode drift (assumed to be linear) was determined at approximately 10-day intervals. The drift was approximately 0.002 *pH* unit/day, and the correction was made to the measured values (Byrne *et al.*, 1988; Chen, 1988a, 1989; Chen *et al.*, 1986).

The CTD-Rosette used to obtain deep samples malfunctioned once during INDIVAT 1, so all 11 bottles were closed at approximately 3400 m at GS 427. Four replicate samples were taken from each bottle. The standard deviation of the pH data (44 points) is 0.0027 pH units (1 σ) which includes random error in both sampling and analysis. The standard deviation corresponds to roughly 1 μ mol.kg⁻¹ in total CO₂ (Chen et al., 1986).

Alkalinity and total CO_2 were determined at $25^{\circ} \pm 0.02^{\circ} C$ with an Apple II-controlled titration cell using a program similar to that of Almgren *et al.* (1977) and Bradshaw *et al.* (1981). These measurements have a precision of $\pm 4 \mu \ eq.kg^{-1}$ for alkalinity and $\pm 5 \mu \ mol.kg^{-1}$ for total CO_2 and were performed within 12 hours of sampling (Chen *et al.*, 1986; Keeling *et al.*, 1988). Duplicate samples from 3 Rosette bottles were taken at 3400 m at GS 427. The standard deviation of the alkalinity data is $1.3 \mu \ eq.kg^{-1}$.

Nitrate was analyzed within 12 hours using the flow-injection method of Johnson and Petty (1983) during INDIVAT 1 (Chen *et al.*, 1986). The precision was $\pm 0.2 \,\mu \, mol.kg^{-1}$. The above mentioned *pH*, alkalinity, total CO_2 and nitrate data are listed in a technical report (Chen *et al.*, 1986) and are not relisted here. Poisson *et al.* (1985) measured and reported nutrients with an autoanalyzer during INDIGO 1 / INDIVAT 3 with higher precision.

4. CHEMISTRY OF THE SURFACE WATERS

Many surface-water chemical properties, especially when normalized to a constant salinity to remove the effects of evaporation and precipitation, are known to correlate linearly with temperature (e.g., nitrate: Chen *et al.*, 1982b; pH, phosphate and silicate:

Chen, 1984; calcium: Chen *et al.*, 1982a; alkalinity: Edmond, 1974; Chen and Millero, 1979; and total CO_2 : Chen and Millero, 1979). My normalized nitrate ($NNO_3 = NO_3 \times 35/S$) values for surface waters are also found to correlate generally linearly with surface temperature (Figure 2; Chen, 1988b) between 22° C and 17° C, except for slight changes in slope near the Antarctic and Subpolar Fronts (approximately 4° C and 13° C, respectively). There is essentially no nitrate above 17° C. The normalized phosphate shows the same trend (Chen *et al.*, 1986). Le Corre and Minas (1983) also observed similar phenomena for nitrate and phosphate in the same general region in summer (March) 1977.

The *pH* also correlates linearly with temperature below 17° C (Figure 3) except for a slight change in slope near the fronts. Note that above 17° C the nitrate is essentially used up. There seemed to be a *pH*-stat between 17° C and 23° C during INDIGO 1 / INDIVAT 3, a phenomenon not observed during INDIVAT 1 (Figure 3). The *pH* seems to increase again with temperature above 23° C.

Normalized alkalinity ($NTA = TA \times 35/S$), and normalized total CO_2 ($NTCO_2 = TCO_2 \times 35/S$) also correlate roughly linearly with temperature (Figures 4 and 5). There may be a slight change in slope near the fronts. A change in slope also occurs at approximately 20° C. I suspect this change is caused partly by equatorial upwelling. The upwelled water has high NTA and $NTCO_2$ concentrations but low *pH* values, causing the slopes to change accordingly.



Fig. 2. Normalized nitrate vs. temperature for surface samples collected during INDIVAT 1, INDIGO 1 / INDIVAT 3, and GEOSECS expeditions (taken from Chen, 1988b).



Fig. 3. pH (25° C) vs. temperature for surface samples collected during INDIVAT 1 and IN-DIGO 1 / INDIVAT 3 expeditions.



Fig. 4. Normalized alkalinity vs. temperature for surface samples collected during INDIVAT 1, INDIGO 1 / INDIVAT 3, and GEOSECS expeditions.



Fig. 5. Normalized total CO_2 vs. temperature for surface samples collected during INDIVAT 1, INDIGO 1 / INDIVAT 3, and GEOSECS expeditions.

It is not clear why such linear correlations exist. I assume that the biological activity reduces the INDIVAT $1 NNO_3$ concentration from $28 \mu mol kg^{-1}$ in the Circumpolar Current near 50° S to $0 \mu mol kg^{-1}$ in the subtropical region near 35° S at a rate linearly correlated with the warming of the water from 2° C to 17° C. Given the C/N ratio of 106/16 (Redfield *et al.*, 1963), this consumption of nitrate must produce 185.5 $\mu mol kg^{-1}$ of organic carbon. At the same time, the NTCO₂ must be reduced by the same amount. In addition, the warming of seawater drives out dissolved CO_2 and reduces the NTCO₂ by $8 \mu mol kg^{-1}$, assuming that the surface seawater remains at the same degree of saturation with respect to CO_2 .

Production of organic carbon as soft tissue is associated with the production of inorganic carbon as hard tissue or shells at roughly a four to one ratio (Broecker and Peng, 1982). Thus, the production of $185.5 \,\mu \, mol \, kg^{-1}$ in organic carbon should result in a further reduction of $46.4 \,\mu \, mol \, kg^{-1}$ in NTCO₂ and $63 \,\mu \, eq \, kg^{-1}$ in NTA after taking into consideration the effect of nitrate and phosphate on alkalinity (Brewer *et al.*, 1975; Dyrssen, 1977; Chen *et al.*, 1982a). Consequently, I expected a total reduction in TCO₂ of 240 $\mu \, mol \, kg^{-1}$ and observed a reduction of $215 \,\mu \, mol \, kg^{-1}$. I also observed a NTA reduction of $70 \,\mu \, mol \, kg^{-1}$. These correlations indicate that biological activities contribute to most of the reduction in nitrate, alkalinity and total CO₂. For waters north of 35° S (waters warmer than 17° C), nitrate concentration is so low that other sources of nitrogen, such as ammonia, or nitrogen fixers may be important in biological consumption. Thus, the Redfield ratio is no longer applicable. Further, the effect of equatorial upwelling becomes important, and simple linear relations do not exist.

The decrease in NNO_3 of $28 \mu \mod kg^{-1}$ between 4 and $17^\circ C$ for INDIGO 1 / INDIVAT 3 corresponds to the decrease of $239 \mu \mod kg^{-1}$ in $NTCO_2$ and $63 \mu eq kg^{-1}$ in NTA. I observed a similar decrease in $NTCO_2$ and NTA of $200 \mu \mod kg^{-1}$ and $75 \mu eq kg^{-1}$, respectively (Chen *et al.*, 1986). These results strengthen the suggestion that biological activities contribute to most of the reduction in nitrate, alkalinity and total CO_2 before the Antarctic waters reach the tropical zone. It should be pointed out that the above is the first attempt to make calculations along this line and it remains to be seen whether the assumptions are valid.

5. CHEMISTRY OF THE SUBSURFACE WATERS

During INDIVAT 1 only limited samples were collected at two stations, GS 427 and 429. The temperature, salinity, pH, NTA and $NTCO_2$ for these stations are plotted vs. depth in Figures 6 and 7, respectively (Chen *et al.*, 1986). GS 427 is north of the Subtropical Front; Antarctic Intermediate Water (AAIW) is found here as a S-min layer at approximately 1000 m. A pH-min is located slightly below this depth. NTA and $NTCO_2$ seem to increase with depth, but at a faster rate near the surface. Most of this increase is due to the increase in preformed values.

GS 429 is south of the Antarctic Front. AAIW is absent here (Figure 7). T, S, pH, NTA, $NTCO_2$ and NNO_3 (not shown on Figure \mathcal{A}) remain constant above 150 m due to winter mixing. To my knowledge, this is the first time such winter data have been collected in the Indian ocean section of the Circumpolar Water (Chen *et al.*, 1986).

A very pronounced *pH*-min occurs at 800 *m*, with a $NTCO_2$ -max immediately below it. They do not coincide, perhaps, because dissolution of $CaCO_3$ below 800 *m*, signaled by the increase of *NTA*, increases $NTCO_2$ further but does not affect *pH*. A very weak S-max exists at about 2000 *m*, reflecting the influence of the remnant North Atlantic Deep water (NADW) which is low in $NTCO_2$ (minimum; Figure 7) and low in nutrients but high in *pH* (maximum; Figure 7). The *pH* and $NTCO_2$ signals are much stronger than the salinity signal and are quite useful as additional tracers in the Southern Ocean (Chen, 1984; Chen and Rodman, 1985, 1990).

The temperature cross-section for the stations occupied during the INDIGO 1 / INDIVAT 3 expedition is given in Figure 8. Upwelling is evident for the southernmost stations (G 10 – 16), and is also shown clearly in the salinity cross-section (Figure 9). In addition, Figure 9 shows the Subtropical Front near 43° S and the low salinity tongue of AAIW. One particularly interesting feature was the core of S-max water found at about 2700 m at G 9 near 43° S. This is probably the core of remnant NADW (Redfield, 1960), characterized by high salinity and pH but low nitrate, phosphate, silicate, NTA, NTCO₂,



Fig. 6. Vertical profiles of temperature, salinity, pH, NTA, and NTCO₂ at GS 427 reoccupied during INDIVAT 1 (T, S data taken from Poisson et al., 1985). The lines for pH, NTA and NTCO₂ are drawn by eye. Note the changes in scales.



Fig. 7. Vertical profiles of temperature, salinity, pH, NTA, and $NTCO_2$ at GS 429 reoccupied during INDIVAT 1 (T, S data taken from Poisson *et al.*, 1985). The lines for pH, NTA and $NTCO_2$ are drawn by eye. Note the changes in scales.

 pCO_2 , NCa (normalized calcium = $Ca \times 35 / S$) and AOU (apparent oxygen utilization) (Poisson *et al.*, 1985; Chen *et al.*, 1986).

The *pH* cross-section is given in Figure 10. Decomposition of soft tissue decreases the *pH* with increasing water depth. However, the water near the bottom is affected by AABW, which has a relatively high *pH* value. As a result, a *pH*-min layer is formed (Chen, 1984; Chen and Rodman, 1985; Chen *et al.*, 1986).

The NTA cross-section is given in Figure 11. NTA increases with depth almost monotonically in regions north of the Subtropical Front because of the dissolution of calcareous hard tissue and shells. South of the Subtropical Front, the water contains mainly



Fig. 8. The temperature cross-section for stations occupied during the INDIGO 1/INDIVAT 3 Expedition (data taken from Poisson et al., 1985).

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Fig. 9. The salinity cross-section for stations occupied during the INDIGO 1 / INDIVAT 3 Expedition. (data taken from Poisson et al., 1985)



Fig.10. The pH cross-section for stations occupied during the INDIGO 1 / INDIVAT 3 Expedition.

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Fig. 11. The NTA cross-section for stations occupied during the INDIGO 1 / INDIVAT 3 Expedition.

siliceous organisms so that the increase in NTA with depth is small. Because Circumpolar upwelling brings deep water (and its high NTA) to the surface, the entire region south of the front is high in NTA throughout the water column. Biological consumption reduces surface NTA again when the water flows northward. Note the NTA-max at G 1 caused by the undercutting of AABW. The data of Naqvi and Naik (1983) and Sen Gupta and Naqvi (1984) in the North Indian Ocean show this NTA maximum clearly.

Figure 12 gives the $NTCO_2$ cross-section. The resolution is coarser than the *pH* plot because of the relatively poor precision of the $NTCO_2$ data. Nevertheless, the major features are preserved, i.e. the core of $NTCO_2$ -min water at G 9 (also shown on S and *pH* plots), the $NTCO_2$ -max at G 3 (also shown on NTA plot), and the effect of Circumpolar upwelling (also shown on T, S, *pH*, and NTA plots). For comparison, the apparent oxygen utilization cross-section is presented in Figure 13 (Chen *et al.*, 1986). The similarity between Figures 12 and 13 is apparent. The AOU cross-section shows a maximum at the southernmost stations. This maximum corresponds to the *pH* minimum found on Figure 10. However, the AOU maximum for the northernmost stations fall slightly deeper than the *pH*-min layer.

6. COMPARISON OF INDIVAT 1 AND INDIGO 1 / INDIVAT 3 DATA WITH GEOSECS DATA

Even when data are collected at the same locations, it is difficult to compare those collected at different times because one can never be sure that the same waters, especially surface waters, have been sampled. For example, surface NO_3 values show significant variations over three cruises to the same location. At GS 429, occupied in summer (Feb. 1978), the surface concentration was $20.2 \ \mu \ mol \ kg^{-1}$ (Weiss *et al.*, 1983). The INDIGO 1 / INDIVAT 3 value was $21.2 \ \mu \ mol \ kg^{-1}$ (March 1985) and the INDIVAT 1 value was $24.5 \ \mu \ mol \ kg^{-1}$ (July 1984). The correlations between NNO_3 and temperature, however, only changed slightly, with the INDIGO 1 / INDIVAT 3 concentrations systematically higher (Figure 2). For waters south of the Subtropical Front, I also do not see much difference between the *pH*/temperature correlation for data collected in winter and that obtained in summer given the statistical accuracy of the data.

There are, however, significant differences in surface NTA and NTCO₂ values among the three cruises (I subtracted 15 μ mol kg⁻¹ from the reported TCO₂ values according to the recommendation given in Weiss *et al.*, 1983). INDIGO 1 / INDIVAT 3 seems to have produced the highest NTA and NTCO₂ values. These NTA values are approximately 5 μ eq kg⁻¹ higher than the INDIVAT 1 values, which are in turn 10 μ eq kg⁻¹ higher than the GEOSECS values (Figure 4). The INDIGO 1 / INDIVAT 3 NTCO₂ values are approximately 20 μ mol kg⁻¹ higher than the INDIVAT 1 values, which are in turn 10 μ mol kg⁻¹ higher than GEOSECS (Figure 5). These differences may indicate either seasonal effects or systematic differences in analysis. More data are needed in order to make a better judgement.

Vertical profiles of NTA and $NTCO_2$ were collected at GS 427 during INDIVAT 1 and during INDIGO 1 / INDIVAT 3 by Chen (Chen *et al.*, 1986) and by Poisson (Poisson *et al.*,



Fig. 12. The NTCO₂ cross-section for stations occupied during the INDIGO 1 / INDIVAT 3 Expedition.



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Fig. 13. The AOU cross-section for stations occupied during the INDIGO 1 / INDIVAT 3 Expedition (data taken from Poisson et al., 1985).

1985) independently of each other, and during GEOSECS. These four sets of data generally agree to within $10 \mu eq kg^{-1}$ for NTA and $10 \mu mol kg^{-1}$ for NTCO₂, variations only slightly larger than the combined experimental error. The agreement is equally good at GS 428 (Chen *et al.*, 1986).

Significant differences, however, exist between different data sets at GS 429. There is a large difference in *NTCO2* near surface, with the winter INDIVAT 1 values the highest (between 20 and 25 μ mol kg⁻¹ higher than the INDIGO 1 and GEOSECS data), obviously because of seasonal effect. The smaller systematic difference for subsurface waters, however, is probably due to analytical errors. Poisson's *NTA* values are approximately 10 μ eq kg⁻¹ higher than the GEOSECS data and both are approximately 10 μ mol kg⁻¹ higher than the GEOSECS data with Chen's data in between the two. Poisson's *NTCO*₂ values agree with Chen's data and both are approximately 10 μ mol kg⁻¹ higher than the GEOSECS data (Chen et al., 1986; Poisson et al., 1985).

7. CONCLUSION

I have obtained the first contemporary winter nitrate and carbonate data in the southwestern Indian Ocean. Surface nutrients, pH, alkalinity and total CO_2 values differ significantly from the summer data when compared at the same location. But the differences are less pronounced when compared at the same salinity and temperature. These winter data can now be used to estimate the initial concentrations of nutrients, pH, alkalinity and total CO_2 for waters formed in the southwestern Indian Ocean, such as the Antarctic Intermediate water in the Indian Ocean sector.

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西南印度洋二氧化碳及营養鹽 之冬夏對比

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摘 要

1984年南半球冬季(七月)及1985年南半球夏季(二、三月)時 ,作者利用法國研究船 Marion Dufresne 號於西南印度洋取得了若干化 學數據。取得之冬季數據代表南極中層水於其一個重要形成地區之 起始值。有了起始值之後,方能研判南極中層水於印度洋流動時所 產生之化學變化。

取得之數據可作冬夏對比,並且可與1978年Geosecs 航次取得之數 據比較。初步結果顧示:表水之pH,及鹽度標準化後之硝酸鹽 (NNO₃),鹼度(NTA)及總二氧化碳(NTCO₂)與水溫間之變化呈直線關 係;偏離直線之部份與次南極鋒面、亞熱帶鋒面、及赤道湧昇流有 關;同一地點之硝酸鹽濃度及 pH 顧示相當大的季節性變化。於相同 水溫處相比,則變化較小;但NTA及 NTCO₂即使於相同水溫時相比, 似乎亦顧現出季節性變化;南極水流至印度洋中部時所減少之NTA 及 NTCO₂,可能能夠以消耗 NNO₃ 及水溫上昇解釋;殘餘之北大西洋 深層水之鹽度信號甚小,不易辨別,但可輕易地由 pH 及總二氧化碳 信號分辨出;營養鹽及溶氧亦有助於追蹤北大西洋深層水。