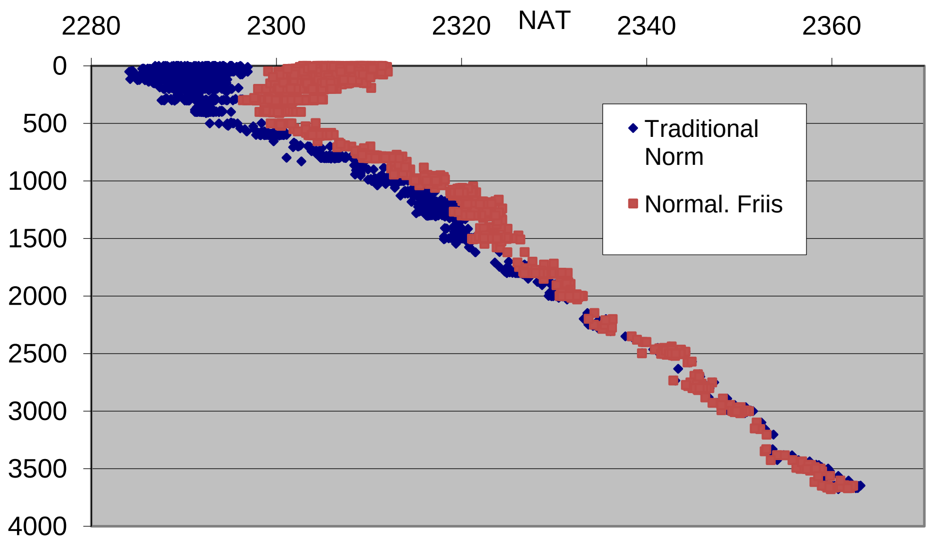
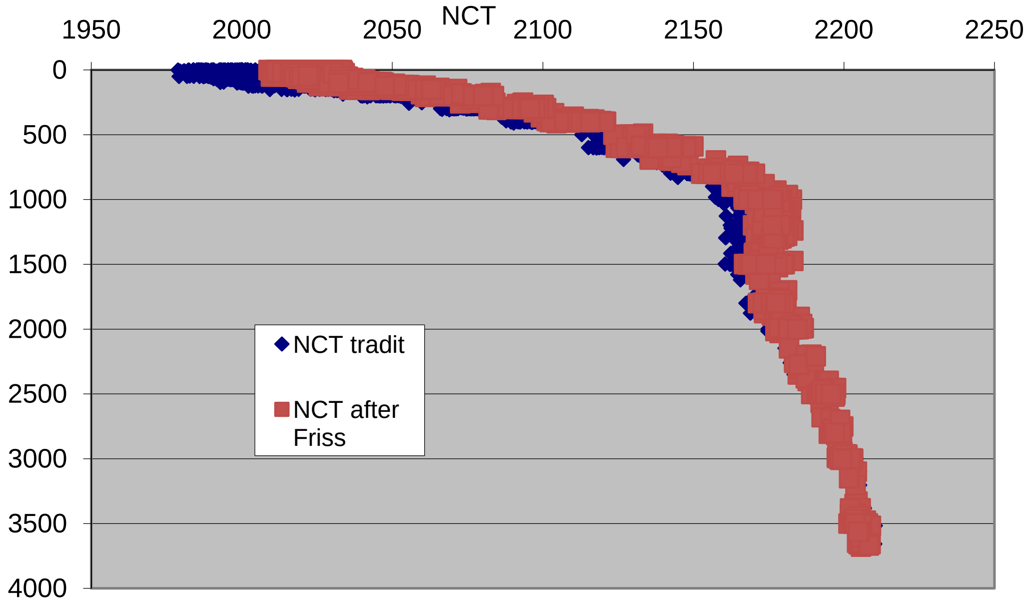
Anonymous Referee #1

First of all, we would like to thank reviewer #1 by his/her work and valuable comments that have improved the quality of our initial manuscript. Most of the indications and suggestions have been included.

Decision: This paper describes important time series observations. It can be published after major clarifications and revision.
General comments
This paper describes 10 years of time series carbon chemistry observations from the ESTOC site in the Sub-tropical N-Atlantic. Time series observations are very valuable at times of increasing atmospheric carbon dioxide when the oceanic response needs to be well documented and understood.
The authors describe the carbon chemistry results both on the basis of their direct observations and they have also chosen to normalize total inorganic carbon, CT, and Alkalinity to salinity=35 (page 1999, line 3). There is no reason given for the normalization. It is evident from Fig. 8 that the observation data covers a salinity range from S<35 to S >37, and that several water masses at various depths are involved. Here the normalization to S=35, the lower end of the observation range, means a substantial change in numeric values. Salinity normalization of carbon chemistry data ignores the effects of dissolution of biogenic carbonates, alkalinity inputs from rivers and may lead to relationships which do not reflect reality (Friis et al., 2003). Direct salinity normalization is really justifiable only when the salinity changes are solely due to evaporation or precipitation of water, i.e. cases where the AT-S relationship has a zero intercept. The authors must discuss the advantages and disadvantages of the salinity normalization if they conclude it is necessary for the interpretation of the observations. The interpretation basis needs to be sound, the present mixture of data and normalized data is confusing and unacceptable.

We have introduced the calculation of normalized values following Friis et al., 2003 (Experimental section) and indications about its necessity. Also, the discussion of data with both normalized and non-normalized data has been reduced, and only some indications are given in order to have data for comparison with other sites and oceanic areas in future works.
As indicated in Friis et al., 2007, the most important variation between traditional normalization and that indicated in Friis et al., 2003 is observed in the first hundreds of meters.





The narration on page 2003 of carbonate system variable distributions with depth and water masses is not vivid. Normalized pH25, NCT and NAT are used, and also unnormalized values, without any advantage being apparent with the normalizations. A graphic + verbal presentation is likely to be more informative. As indicated before, some parts have been reduced or removed.

The description, on pages 2005-2006, of variations in the rate of carbonate system changes with depth is based on 12 fixed depth levels. This is supplemented with Table 1 and figures 4, 5, which are crowded, and 6. The number of levels is superfluous, the uppermost 3, in the seasonally mixed layer, show essentially the same trends. It would be more interesting to present the surface layer and then base the description of the lower levels on hydrographic properties, water masses. Indeed, the authors venture into this path when they note NADW changes on page 206, line 23. Here again the advantage of normalization is not clear, if any. Table 1 includes both normalized and unnormalized data. The carbonate system species are calculated from normalized pH25 and alkalinity as observed, page 2007, line 17.

We agree with the reviewer that values in the first 100 m show similar trends. Following his indication, figures 4 and 5 include now a value for the mixed layer. However, in Table 1, we consider that keeping the full values can be useful in any future work and for that we have kept and introduce the value in the mixed layer. As seasons are covered in a time series station and MLD varies along the year, we have computed accordingly the trends along the series of data.

Section 3.3 presents interesting results on anthropogenic carbon estimates. Firstly an evaluation using the TrOCA method, presumably on unnormalized data.

That is right.

Secondly, on page 2013, a comparison of the TrOCA results with the rate of CT and NCT increase with time. The normalization question comes up here once more. The argument is based on that "oceanic and physical conditions are constant" (page 2013, line 4) and relatively constant potential density is invoked for the fixed depth levels. This appears to be unrealistic assumption, the reality is stated on page 2015, lines 9-12.

Really the argument indicated was only considered when we wanted to extrapolated our data for other years. As we do not see changes in temperature and salinity along the years below the MLD, this assumption can be considered and will allow us to follow biochemical changes.

A new abstract will need to be written after the revision.

The abstract has been updated.

Specific comments

P1996, line 22: The reference Bates, 2007 seems appropriate here.

We have changed the text.

P1999, line 7: The Experimental section should give a clear picture of the methods and procedures applied to ensure consistent quality information over the period of the time series. Thus information on precision evaluation and assessment of accuracy should be presented. The presentation here is unclear in many instances as listed in the following comments.

We have changed the experimental section considering this and next comments

P1999, line 10: Fig. 1 would be more informative if the ESTOC location was shown on a chart of surface currents.

Figure 1 has been changed to a new one

P1999, line 11: Description of the work conducted by German partners seems irrelevant if these observations are not used in this work.

It was removed.

P1999, line 20: Data and results are presented from depths >1000m, so "the first 1000 m" must be an inaccurate description.

We have changed and explained better the data considered in this study.

P1999, line 24: Needs fuller information on CTD-SIS. Which CTD and what does SIS stand for?

It has been changed

P2000, line 1: How was the salinity determined in the water collected in Niskin bottles?

It was included

P2000, line 5: One would expect information on discrete fCO2 sampling, or was fCO2 only determined in the surface water flow system?

fCO_2 was only measured in surface data, as indicated.

P2000, line 9: The standard deviation of 0.0015 is mentioned for determinations of

CRM Batch #42 which was issued in 1995. Dickson Water is not certified for pH. Is this intended to be information on precision or accuracy?
It was considered and explained.
P2000, line 13: CRM #32 was issued in 1995, #35 in 1996, #42 in 1997 and #46 in 1998. What QC procedures were applied for alkalinity 1998-2004?
It was considered and explained.
P2000, line 14: "values that were within _1.5 _mol kg-1". Is this precision or accuracy?
It was considered and explained.
P2000, line 18: What does the "residual of _3 _mol kg-1" stand for. If it is the difference between calculated and CRM concentrations, then were corrections applied? How was the period 1998-2004 treated? Considering the precisions of the pH and Alk determinations what is the uncertinty associated with the CT calculation?
It was considered and explained.
P2000, line 20: Were CRMs used with the CT determinations after 2004?
Yes. It was considered and explained.
P2001, line 6: What is the "zero reading" gas?
It was considered and explained.
P2001, line 12: Does the VOS-QUIMA line pass the ESTOC location?
It was indicated in this new version
P2001, line 16: Please clarify "standard error" for oxygen and "standard deviation" for nutrients.
It was considered and explained.
P2005, line 1: The treatment on surface water fCO2 is excessively long and the conclusion on rising surface fCO2 trend is not convincing, in particular because the uncertainty for the 1995-2004 trend, 1.55 _atm yr-1 (page 2004, line 6), is not given. What can be deduced about this trend from the air-sea fCO2 difference?
It was considered and explained.
P2006, line 5: Explain the calculation "fifth of the annual increase: : :".
It was an error in our expression.
P2006, line 7: Why assume alkalinity when measurements exist?
It was considered and changed.
P2007, line 9: It seems appropriate to compare this rate with other observations from the Sub-tropical N-Atlantic, e.g. (Bates, 2007).
It was considered
P2007, line 12: The effect of temperature on the carbonate equilibria is well known but this comparison should provoke thoughts on the necessity of pH temperature normalization.
We have included some comments about the differences observed.
P2007, line 28: Calcium carbonate saturation is affected by pressure, fCO2 and and temperature. Is the change in omega values only due to decrease in carbonate concentration?
We agree with the reviewer and it has included.
P2008, line 7: If the authors can show that the ".. rate of change remains constant.." is a realistic model both on grounds of CO2 emissions and sea water carbonate chemistry then the calculations that follow are presentable, otherwise not.
We only assume that the trend presented here can be extrapolated to this century. Taking into account the increase in CO2 emissions and effects on the ocean chemistry

and ocean sequestration, this can be considered an estimation of future evolution and can be considered as a minimum value.

P2009, line 9: Section 3.2 is interesting. It appears from figures 9 and 10 that the profiles are created from mean results. The authors must reveal how the time series data are used in this section. Secondly, the questions on salinity normalization in General comments come up here again.

We have included a description how the data were computed.

P2010, line 11: How does ATorg become NA Torg ?

It was indicated how it was calculated (first it was first considered on AT and then normalized).

P2010, line 25: How does RC . AOU become NC Torg ?

It was indicated how it was calculated (first it was first considered on AT and then normalized)

P2013, line 13: The argument for "increasing storage capacity" is not convinging. How do the authors define "storage capacity" and for what reason is it increasing?

We have changed that in this new version

P2016, line 2: Is the statement "general concordance : : : with others in the area" a comparison with the BATS time series? If so, then reference is needed.

The comparison was done in the discussion of the data. In the conclusion we have not included any reference.

P2016, line 3: The final sentence states that the ESTOC site can be used by way of a reference to monitor anthropogenic carbon changes in the N-Atlantic Sub-tropical gyre. This statement cannot be presented without a substance. What are the criteria for reference stations which ESTOC supposedly fulfills? Have these criteria been put forward by international agencies?

We have changed our indication.

Technical corrections

P1996, line 4: "anthropogenic properties" is a strange concept, seems better to use "anthropogenic signatures".

Done

P1996, line 10: "experimental values" better: "observed values"

Done

P1998, line 13: "experimental" better: "observed"

Done

P2001, line 20: Sectioin 3.1 is far too long and could be broken up by subjects.

Done

P2003, line 10: Is 2010 a typo, should it be 2100?

Done

P2004, line 1: Suggest new subsection

Done

P2005, line 6: Suggest new subsection

Done

P2010, line 26: Typo: hard-issues

Corrected

P2023: Fig. 2 The figures are small and of low resolution, the numbers are illegible even after zooming in.

A new set of figures has been included.