Reply to G. Wiesenberg

We thank the referee G. Wiesenberg for his comments and questions on our manuscript. Below we have provided point to point responses to each referee comment/ question (in italic).

A very basic comment to a general point of the reviewer´s criticism is provided here as an introduction to our reply. The well-intended suggestions to include more parameters for determination of physical, chemical or biological properties are one key element in the criticism of the anonymous reviewer and also of reviewer G. Wiesenberg. We fully agree with the reviewers that analyzing more parameters will significantly improve understanding of soil heterogeneity and as such is highly desirable but obviously, time and budget limitation prevented us from including more parameters. We have tried to keep the selection of biogeochemical parameters used for our assessment of paddy soil heterogeneity as broad as possible, including highly reactive and more conservative properties. We furthermore attempted to include some of those parameters that are frequently used in other biogeochemical studies without claiming to cover all aspects of soil biogeochemistry. It is our hope that this approach will be appreciated by the readers.

The current manuscript determined inter- and intra-site variability of organic matter properties in a chronosequence of paddy soils in China. Mainly intra-site variability related to a different time of rice cultivation was found to sustainably change 'conservative' soil organic matter properties. Also 'rapid' changes were observed to increase TOC contents in paddy soils within the first 300 years of rice cultivation. Afterwards, biogeochemical parameters do not undergo any further substantial change. This study is based on a sophisticated sampling design, using site and field replicates and application of various statistical approaches to determine clustering of individual sites. The combination of molecular, elemental studies with plot-related and field replicates has not been applied frequently and thus provides scientific innovations in that sense. However, the argumentation in many points and discussion of data with respect to available literature is not adequate in the current version of the manuscript. I.e. the reader of the manuscript might get the impression that numerous of the investigated parameters have been analysed for the first time or how the data is related to available literature (if there is some available). In some parts of the manuscript mistakes were observed and in other parts the meaning of the study is discussed in a very enthusiastic way, leading to conclusions, which are not completely supported by the data. Hence, some modifications are required before accepting the final version of the manuscript. But due to the overall improvements in combing several orders of scales, I would recommend acceptance of the manuscript after moderate changes.

The scope of this study was to investigate the biogeochemical heterogeneity of paddy soils and for this purpose we selected a manageable suite of different parameters that we assumed will vary either to a minor or major extent in space and time. A discussion of our "heterogeneity" data with existing literature on this issue is severely limited because, to the best of our knowledge, we could not identify relevant publications for paddy soil systems. This by no means shall indicate that individual parameters have not been analyzed in paddy soils before but in the combination that we present we are not aware of any comparable study.

(1) *Although it was not the main focus of the manuscript to simply determine SOM properties, a minimum of discussion is required, how the observed data fits literature results. This is completely missing and should be added.* **(2)** *The definition of 'conservative' vs. 'non-conservative' parameters is not clear. How can the authors group some of the parameters without using any relevant literature arguing for their selection. Although this grouping seems to be logical in many points, it has to be supported by literature data. Even if such literature is not available for paddy soils, some data (^{14}C dating and/or turnover times) should be available for other soils and sediments.* **(3)** *At several points of the manuscript the 'minerogenic composition' is discussed to have an influence on the inter- and intrasite variability of SOM. Of course this is one potential factor, which somehow has to be confirmed. Otherwise such statements remain speculative. Hence, it should be indicated, whether this is proven or 'likely' as other factors cannot completely explain individual observations.*

Ad (1): The focus of our study was to assess the intrinsic heterogeneity of paddy soil biogeochemistry and as such we have not compared our "heterogeneity" results with literature data, simply because to the best of our knowledge such data does not exists. A detailed comparison of biogeochemical heterogeneity as such is thus not feasible. In order to put our field "averages" into context with other investigations we have added several literature references.

Ad (2): The definition and differentiation of "conservative" and "non-conservative" biogeochemical parameters/properties, is to a certain extent intuitive or logical as we have expressed in the relevant section of the manuscript. We have added some literature data to this section: As labile parameters microbial biomass carbon and nitrogen (C_{mic}, N_{mic}) (Refs: Shibahara et al., 1995; Chantigny et al., 1996; Bai et al., 2000; Bannert et al., 2011) nitrate, ammonium (Refs: Myrold et al., 1986; Davidson et al., 1992; Stark, 1997), dissolved organic N, and dissolved organic C (Kalbitz et al., 2003) were considered. Figure 3 demonstrates that our differentiation into "conservative" and "non-conservative" parameters is fully supported by the coefficient of variation values obtained from statistical analysis.

Ad (3): We have used paddy soil spectral colour values and magnetic susceptibility as proxies representing changes in minerogenic composition, in particular in redox-dependent iron species. A detailed mineral analysis including iron oxide/oxyhydrate coatings was beyond the scope of this study. Our data show that using the susceptibility and colour proxies a clear separation of all field sites is achieved (see figure 5b), with inter-site variability exceeding intra-site variability (as shown in table 2 by Kruskal Wallis test).

Abstract Line 17ff The lowering in CV from younger to older sites could be also attributed to degradation of some parts of OM and not only the longer usage. Paddy fields that are used for several hundred years should be more or less under steady state conditions, which argues against duration of usage as single effect.

Degradation of soil organic matter will ultimately lead to a more homogeneous "residue" and thus the comment by reviewer G. Wiesenberg is correct. This degradation will occur in parallel to utilization time and hence our statement is correct as well. The fresh addition of crop residues, root exsudates, manure and fertilizers according to different management practises over time argues against a steady state system in the paddy soils analyzed (see Bannert et al., 2011a,b or Roth et al., 2011).

1. Introduction In the introduction section, a consequent literature review of a couple of factors is simply missing. Numerous statements and assumptions have to be supported by adequate literature. Only few examples are given below. In general, the section has to be strongly improved in that context. Page 4 There should be some adequate literature be available, which supports the statements given in the introduction as e.g. the conservativeness of parameters like TOC, isotope and lipid composition, as well as for assumed labile parameters. This literature should be added. Similarly, the factors influencing soil heterogeneity should be supported by literature.

We have added references but it should be realized that such literature references for paddy soils developed on marine tidal flat substrates are not as frequent as may be expected. Comparison with other paddy cultivation systems is of limited use and adoption of literature data obtained from studies of grassland, forest or upland cropping systems may be completely misleading. We have thus kept literature references to a minimum.

2.1 Study sites Page 6 As WRB was used for soil classification, soil names should be written according to WRB guidelines, i.e. with large letters for the main soil groups.

Corrected

2.2 Sampling Page 6 It is unclear, how samples were collected, i.e. using a spade or an auger ('roughly 20 cm' is not precise enough in that sense). This is of special importance as it should be clear, whether the sample is representative for the whole sampling interval (auger) or is just a 'mixed' sample, which could yield more top soil than deep soil material (spade).

The sampling system consisted of an auger as depicted in Fig. 2 and a metal spacer used to obtain 7 individual field subsamples at identical spacing. The samples were all collected from the puddled horizon and did not include material from the plough pan. As the fields are puddled twice per year there is no difference between top soil or deep soil.

2.3 Laboratory analyses Pages 7-8 In Chapter 2.2 it was stated that samples were freeze dried and ground to fine powder. Is this also true for the samples amended to fumigation for soil microbial biomass estimation? If not, this should be indicated. But if it would be the case, one can doubt about the outcome of the analyses. Remove 'hydrocarbon' in line 23. Add static time for ASE extraction and provide details regarding GC-MS temperature program.

Methods were described in more detail. Fumigation was done on fresh and non-dried soil samples.

3.1 Soil parameters Pages 8-9 Chapter 3.1 is completely redundant. It just repeats parts of the M&M section without providing new information. Thus, this should be removed.

We provided this very short paragraph as an introduction into the discussion section to improve readability. If, unfortunately the opposite effect was achieved we remove these sentences.

3.2 Macroscale intra-site variability Page 9 It remains completely unclear, how the authors could attribute the individual parameters as 'conservative' or 'non-conservative'. This must be supported by adequate literature and should be also added in the text body, but not only in Table 1.

The definition and differentiation of "conservative" and "non-conservative" parameters is given in the introduction at end of page 3 beginning of page 4. As mentioned above, literature data for paddy soil developed on marine substrate is scarce and thus only a few references have been added (see response to point 1).

3.2.2 Lipid and alkane conc and comp The authors state that alkanes contribute to total lipids only a rather short portion, whereas other lipids are much more prominent. So, why were the other lipid classes, which were also achieved during extraction (but probably not during separation), not analyzed? Analyses on terrestrial soils identified substantial differences in the stability and cycling of lipids in soils. Additionally, incomplete combustion has been described to sustainably contribute to specific alkane patterns in soils. In the current version of the manuscript it is questionable, whether these are assumptions in the current study, proven by comparison with literature results or simply speculations. The authors could simply face this by including relevant literature, which in major parts is completely missing.

As mentioned several times above, we do not consider the various literature results determined on upland cropping systems, grassland or forests as directly transferable to paddy soils, which as temporarily subaquatic soils with highly alternating redox states behave differently. Thus we have kept the comparison with standard literature to a minimum. Furthermore, the aim and scope of this paper is not the comparison of paddy soils with non-paddy soils but the assessment of heterogeneity within paddy fields. Thus our comparison is between our study sites and not between our sites and others. Literature references in this respect are consequently of limited value.
As mentioned in the introduction, we had to make a compromise in the feasibility of this study. Either concentrate on lipid variability and ignore other parameters or restrict lipid analysis to one compound class. We opted for the second approach and included various other soil biogeochemical parameters. Amongst the lipid classes the n-alkanes are the most reported fraction and hence we selected this class for further analysis as it is most relevant for paddy soil heterogeneity assessment.
Specific n-alkane pattern with an even over odd predominance of short chain homologues was described in diverse studies but was not observed in our topsoils and sediments. Therefore, we do not address this issue in the manuscript.

3.3 inter-site variability Page 17 In Line 11 a 'significant change' was observed. This should be supported by the analyses of statistical siginificance.

Quote. "Contamination of the P700 site did not lead to a significant change in time-integrated basic soil biogeochemical parameters but preferentially affected the aliphatic hydrocarbon composition." There must have been a misunderstanding; we did not observe a significant change in P700 except for aliphatic concentration and composition.

3.4 Organic matter accumulation and sequestration trends Pages 18-19 This chapter in parts sounds a bit strange and should be re-written as some mistakes are included. - When referring to C sequestration and the IPCC report 'rapid' increase in TOC observed in the chronosequence (100 %) is not that surprising and common, whereas e.g. there are numerous examples, where cropping management or cropland to forest conversion can also contribute to similar carbon accumulation (or even more). Controversially, other conversions have been shown to reduce TOC in soil by 50% in less than 10 years. Hence, the manuscript in the current version suggests that rice production could contribute to sustainably sequester carbon in the short term (when using the word 'rapid'), which is not true. This should be changed. - The conversion of soils to rice cultivation cannot really taken into account for counter-balancing atmospheric CO2 increase. numerous FACE experiments clearly showed that biomass production is not strongly promoted under rising CO2 and also TOC is not strongly increasing, rice production cannot solve this problem. In the current version the reader could get this impression, which is also not correct. As Hence, some modifications should be done. - Marschner et al (2006) should be changed to Marschner et al (2008). - The statement of lipids being less recalcitrant than TOC is not true and not supported by the whole literature. In Marschner et al (2008) also alkanes have been described to be more recalcitrant than TOC, depending on cultivation. Additionally, some more recent and also older studies are available, showing that especially alkanes can be more recalcitrant than TOC (e.g. Bol et al., Wiesenberg et al., Feng et al.). It is speculated how n-alkanes are generated in soils, whereas there is numerous literature is available on this topic, which should be cited.

It has not been the intention of this manuscript to develop a full ecological budget for rice cultivation. This is rather impossible as we have studied a very specific setting, i.e. paddy rice cultivation on reclaimed tidal flat sediments in a geographically constrained region in South-East China. The contribution of the rice fields in the Zhejiang Province developed on reclaimed land with only app. 300.000 ha compared to the total cropping area for paddy rice of 160 Mio. ha (Jahn et al., 2012) is negligible. Therefore, conclusions on global CO_2 sequestration via paddy cultivation based on this study cannot be drawn. Under the specific circumstances of the study area a very significant increase in soil organic carbon was observed for top-soils in paddy fields, whereas upland cropping systems in the same region did not reveal a strong increase in top-soil organic carbon (Cheng et al., 2009; Wissing et al., 2011; Jahn et al., 2012). As shown by the data presented here, the increase in topsoil from <0.5 % TOC in tidal wetland to >2.0% TOC occurs in less than 300 years, which to our opinion and according to Targulian and Krasilnikov (2007), is a "rapid" change. It is not the aim and scope of this paper to compare the carbon sequestration rates with other settings that bear completely different boundary conditions.

In this contribution we rather addressed the question that "The chronosequence studied here offers the opportunity to evaluate CO_2 sequestration in paddies, comparison with a non-paddy site (P500) and interferences via intentional management (P1000) or unintentional contamination (P700)" exclusively for our study area.

We have corrected the incorrect reference to Marschner et al. 2008.
In this contribution we have not addressed the generation or turn-over mechanisms of alkanes in soils but rather used the n-alkanes as one representative class of soil lipids. The alkane distribution in this investigation guided in the detection and interpretation of soil disturbance processes. In particular the alkanes revealed a petroleum contamination at site P700 undetectable by other techniques applied.
The paddy top-soil gain in TOC from tidal flat substrate to 300 and 2000 years of cultivations was 460% and 580% respectively (see data in table 1). The gain for total lipids was only 150% and 220%, respectively but the gain for n-alkanes with 350% and 595% was equal to that for TOC. For the specific study site, in terms of stability and accumulation rate the n-alkanes and the total organic carbon appear equivalent whereas the total lipids are more labile. This observation is in agreement with previous studies on non-paddy ecosystems (e.g. Wiesenberg et al., 2004; Marschner et al., 2008). Only under specific circumstances, e.g. in grassland soils, the n-alkanes may even exceed the total soil organic matter pool in stability (Wiesenberg et al., 2008), compensated by an enhanced lability of the carboxylic acid lipids. This clearly indicates that in general n-alkanes are slightly less stable than total soil organic carbon though environment specific exemptions may occur.
We have extended this section in the revised manuscript and added the relevant literature though in principle we do not intend to focus this manuscript onto the subject of soil organic matter and compound class stabilization in soils.

4 Conclusions Page 20 The environmental/ecological budget is not really supported by the data or is only related to a strongly degraded environment before conversion to paddy fields. The last paragraph of the conclusions is not really supported by the data as no real reference soils are regarded. Hence, the last paragraph should be modified in a way that could not lead to the conclusions that paddy soils are a global solution for carbon sequestration. As BIOGEOSCIENCES is a globally distributed journal, such statements as included in chapters 3.4 + 4 could lead some policy makers to the mentioned conclusions.

As mentioned in response to 3.4. we have studied a highly specific environment, reclaimed land from tidal flats that has been converted to paddy rice cultivation. The results obtained from this specific environment cannot be generalized for paddy rice cultivation in other environments. This is emphasized in the revised version of the paper in order to avoid any misunderstandings or misinterpretations.

Fig. 4 ‘tricyclics’ should be changed to ‘tricyclic hydrocarbons’

modified

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