

Dear Edouard Metzger, we would like to thank you a lot for reviewing our manuscript and your constructive comments.

In this response letter *referee comments are listed in italic font*, followed by the authors responses to each individual comment. Changes that will be made to the manuscript are underlined.

Referee comment 1:

Abstract L41-42 I am not sure to follow the causality between metal relative metal solubility and spatial-temporal heterogeneity L45-47 the last sentence of the abstract tends to suggest that decrease of cadmium solubility due to sulfide increase consequently to oxygen declining may affect marine ecosystems. The sentence appears to me overstated, out of the scope of what your data can say, overall, as a general sentence that would appeal to emotion. I would suggest to find another sentence to take perspective. Maybe adding another metal that you must have analyzed using a ICPMS to the study showing different sulfide affinities would have helped to strengthen such a statement.

Response to referee comment 1:

We are using the elements Fe and Cd as role models, as they have a contrasting sulfide solubility, and interpolate their behavior to trace metals which have an intermediate sulfide solubility (lines 86 – 92, 686 – 689, 699 - 701). The concentration/reservoir of a trace metal in the ocean is dependent on the respective input and output fluxes of this element. Consequently, benthic recycling and burial processes are an important aspect of the oceanic mass balance. As especially Fe and also other trace metals can often be the limiting factor for primary productivity, the availability of trace metals has a direct impact on phytoplankton growth. Where bottom water is upwelled to the surface ocean, the benthic burial or release of a trace metal can limit or fuel primary productivity. We are currently working on a second paper where we will present aspects on sedimentary cycling and benthic fluxes of other trace metals off Peru including Zn, which has a low sulfide solubility and Co which has a high sulfide solubility, that are in agreement with the conclusions of the present manuscript.

Referee comment 2:

Introduction All along the introduction, it is stated that little is known about TM benthic fluxes in hypoxic environments. I would disagree with that. The authors cite themselves several of them and I think of studies done by Faganeli and coworkers in the Adriatics, work done during the Microbent project in Thau lagoon including by myself. Work in hypoxic estuaries such as the Gironde or the Scheldt among others. I have the feeling that if authors had detailed more about TM behavior in different environments during occurrence of hypoxia/anoxia and sulfide release into the water column they could have made a series of predictive hypotheses on what would be Cd behavior in the Peruvian OMZ. Overall, I am not a fan of the outline of the introduction because the

rationale is at the beginning and the state of the art shown after is too far to make the reader quite sur about what are really the hypotheses made here and what is really new.

Response to comment 2:

It was not our purpose to disregard earlier work on sedimentary trace metal dynamics in inner-shore and coastal environments like lagoons and estuaries. In sections 1.2 and 1.3 we refer to earlier studies on benthic trace metal biogeochemistry and we will include further citations to studies in inner-shore environments such as Metzger et al., 2007 and Point et al., 2007. We note, however, that except for Fe, little work has been done in open marine systems where the redox- and sediment-dynamics are different compared to estuaries and lagoons. The expansion of our knowledge on trace metal cycling and fluxes in these open-marine environments is the goal of our study.

In the general section on trace metal behavior (1.1 Scientific rationale) we will refer more explicitly to the knowledge gained in studies on estuaries and lagoons that trace metals can precipitate as sulfides when redox conditions change and H₂S concentrations increase.

Referee comment 3:

Methods Incubation time was of 32h, how oxygen evolved within chambers that were not anoxic in the beginning?

Response to referee comment 3:

The evolution of oxygen concentrations during the benthic lander incubations will be published by Clemens et al. (in preparation).

The incubated bottom water in chambers below the oxygen minimum zone (station 9 and 10) did not become anoxic. At station 9 (750 m) the oxygen concentrations decreased from around 3 μmol at the beginning of the incubation to around 2 μmol at the end of the incubation. At the station 10 (950 m) oxygen remained above 10 μmol throughout the incubation time. For the incubation at station 1 (75 m) we cannot exclude that traces of oxygen were present during the first 5 hours of the incubation (some air bubbles may have been trapped inside the chamber during the lowering through the water column, that then dissolved during the beginning of the incubation). This may have reduced dissolved Fe concentrations in the first sample of this incubation through oxidative removal, however excluding this first value would not affect the overall flux magnitude derived from this incubation.

Referee comment 4:

How the benthic boundary layer's thickness is established and what is its dynamics? That should be mentioned in the introduction as how it may affect benthic fluxes What was the detection and quantification limit for Cd measurements

Response to referee comment 4:

It was our goal to sample and to capture the cycling of trace metal cycling in the highly dynamic layer overlying the seafloor. The common definition of the benthic boundary

layer is “a discrete layer of flowing sea water above a benthic substrate, delimited vertically by its contact with free stream flow” (D.J. Wildish, in Encyclopedia of Ocean Sciences (Second Edition), 2001). We acknowledge that, strictly speaking, our sampling interval does not correspond to the benthic boundary layer in the proper sense. We therefore decided to refer to “bottom-near water column” in the revised version.

For Cd measurements we applied the method by Rapp et al., 2017, where the detection limit for Cd is 0.8 pmol L⁻¹, which is well below the lowest concentrations of our samples (40 pmol L⁻¹). The detection limit of Cd will be listed in section 2.2 Analytical methods.

Referee comment 5:

I am aware that there is plenty of literature with diffusive flux calculations from overlying water and the first porewater concentrations. I think this is wrong as soon as a 2-point calculation is subject to the precision of those points and overlying water concentration is an average of 10 to 30 cm of water column according to the fullness of the core. If any author took the entire porewater profile to make an averaged concentration for gradient calculation nobody would take the calculation seriously. I would at least be skeptical about calculations taking only two points to model a line from which, one is an average of something it is impossible to fully describe with such sampling method. I would say that this can be overtaken adding a supplementary point for the gradient determination. Unfortunately, only one of your profiles can apply. This aspect must be clearly discussed in the manuscript and conclusions made from such calculations carefully done. High resolution methods exist.

Response to referee comment 5:

For the diffusive flux calculations we deliberately decided to use the two point concentration gradient between the uppermost pore water sample and the overlying bottom water. We know that this method has its limitations, but need to stay with this approach for the following reasons.

1. This is a commonly used approach which allows us to compare our diffusive fluxes to fluxes from other publications (e.g. Sundby et al., 1986; Warnken et al., 2001; Turetta et al., 2005; Pakhomova et al., 2007; Noffke et al., 2012; Lenstra et al., 2019).
2. The strong gradients between the uppermost pore water and bottom water sample is typically observed within open-marine oxygen minimum zones (e.g. Noffke et al., 2012; Scholz et al., 2019). Since H₂S accumulates close to the surface in these settings, the dissolved Fe peak in pore water is very narrow (order of 1 - 2 cm) and located close to the sediment-water interface. More advanced curve fitting methods would fail to capture sharp gradients at the sediment surface and would, therefore, yield erroneous flux estimates (see also Shibamoto and Harada, 2010; Dale et al., in prep.)

We will explicitly include the above explanation in section 2.3 Flux calculations.

Referee comment 6:

Linear regressions with standard deviation of the slope are not shown in the document, this should appear in the graphics with also error bars for each point

Response to referee comment 6:

We will plot the linear regressions for the concentration change over time during the benthic chamber incubation for Fe and Cd (figures 5 and 7). We will include the coefficient of determination for each linear regression. In figures where error bars are not shown, all symbols are within error as stated in the figure captions.

Referee comment 7:

Particulate flux calculations and CdS water column uptake calculations should also be detailed here, especially the last one

Response to referee comment 7:

The calculations used for the quantification of the sedimentary Cd sink and the different delivery pathways to the sediment are explained in section 4.2.2 Quantification of the sedimentary Cd sink. How we determine the particulate delivery of Cd to the sediment via CdS precipitation in the near-bottom water column is explained below table 3 (Cd enrichment in the sediment after subtraction of diffusive and organic Cd sources). We will also mention this more explicitly in the main text of section 4.2.2 Quantification of the sedimentary Cd sink.

Referee comment 8:

I think that section 3.1. appeals a lot to literature to be part of the result section. Only bottom oxygen data is described from figure 2. I would suggest authors to discard the figure 2, table 2 is sufficient, and put that description in the methods sections For a result section I would avoid naming sections using processes such a “biogeochemical cycling”. I would suggest to simply call it: “Porewater and benthic fluxes” then “iron, then “cadmium”

Response to referee comment 8:

In section 3.1 we present our measured data of oxygen, nitrate and nitrite during both cruises. The prevailing biogeochemical conditions are important for the trace metal cycling- That is why it is crucial to leave this data in the manuscript. Only regarding the very special oceanic condition, the occurrence of a coastal El Niño during our cruise in 2017, we refer to literature instead of our own data as this includes atmospheric and oceanographic observations that we could not conduct ourselves.

We fully agree to change the titles of the results sections as you suggested in 3.2 Bottom water, pore water and benthic flux data and then in subsections 3.2.1 Iron and 3.2.2 Cadmium.

Referee comment 9:

I would also suggest to avoid citations. Why citations are provided for iron and not for

cadmium? Some symmetry should be maintained between these elements at least here to underline differences in results

Response to referee comment 9:

We will remove the first part of the sentence starting in line 354 including the citations in line 355 -356.

Referee comment 10:

In the methods section I made a comment about the thickness of the benthic boundary layer. According to the 4 metres profile from the bottom, it appears that the benthic boundary layer is clearly thinner than 0.5 m as concentrations are stable in your data for almost all profiles. I believe from in situ data of oxygen profiles that this boundary layer is within the range of few micrometers to few centimeters. I am not sure those data are really relevant here and could be discarded or remain as supplementary data. Line 351 authors say that initial incubation concentrations were higher than “bottom samples”. This pleads to the fact that those bottom samples did not describe the benthic boundary layer. This should be said somehow Did the authors thought of showing modelled gradients in the porewater profiles instead of showing modelled incubation points from diffusive fluxes?

Response to referee comment 10:

As mentioned in response to referee comment 4 we will replace the term ‘benthic boundary layer’ by the ‘bottom-near water column’.

Regarding our statement in line 351 (comparing Fe concentrations in the bottom waters collected inside and outside of the benthic chambers): Different Fe concentrations inside and outside of the chamber are to be expected because of diffusive Fe release from the sediment and an accumulation in the enclosed water volume inside the benthic chamber. Furthermore, the bottom water enclosed in the benthic chamber is much closer to the seafloor compared to the bottom water samples collected closest to the seafloor (at 0.5 m). We assume that the concentrations in the bottom water enclosed in the chamber represents the average concentration over the first 30 cm above the seafloor. Outside of the chamber, a concentration gradient between the seafloor and the bottom-near water column is likely to establish because of diffusive release of Fe from the sediment and removal in the bottom water. Therefore, it is not surprising that different concentrations are observed in the chamber and in the bottom water 50 cm above the seafloor.

Furthermore, the water sample at 0.5 m away from the seafloor was collected in sampling bags with peristaltic pumps over a time span of 32 hours. This means that, in contrast to the benthic chamber, where samples were taken within minutes at discrete points in time, short-term concentration changes cannot be seen.

The above explanations will be added to the discussion in section 4.1.1 Comparison of diffusive and in-situ benthic chamber iron fluxes.

As mentioned in response to referee comment 6 we will plot linear regressions for the concentration change over time during the benthic chamber incubation for Fe and Cd

(figures 5 and 7) together with the slopes calculated from pore water profiles to facilitate the comparison of benthic fluxes derived by these two different approaches.

However, applying a transport-reaction model, which would need to involve carbon, sulfur, nitrogen and iron turnover to generate accurate pore water profiles of dissolved Fe, is beyond the scope of the present paper.

Referee comment 11:

4.1.1. Talking about incubations 1, 9 and 10 you mention bioturbation as a potential artefact, what about oxygen consumption and sulfide precipitation during incubation? Actually a combination of all that could happen as oxygen depletion could bring endobionts to surface (see riedel et al experiments in the adriatics) or enhance bioirrigation (Duport et al, 2007) favoring sulfide efflux or iron precipitation. Did you consider leaks in the chamber or dysfunctional homogenization? This is a current issue with in situ incubation. Oxygen and sulphide data from incubations are really missing here to state about ventilation processes or leaking

Response to referee comment 11:

Hydrogen sulfide could not be detected within bottom waters from benthic chamber incubations with our method, which has a detection limit of $1 \mu\text{mol L}^{-1}$. For Fe to precipitate as sulfide several hundred $\mu\text{mol L}^{-1}$ of H_2S are required. Therefore, we exclude Fe sulfide precipitation within the benthic chambers.

As mentioned in our response to referee comment 3, we cannot exclude a slight oxygen contamination within the benthic chamber at station 1 during the very beginning of the incubation (some air bubbles may have been trapped inside the chamber during the lowering through the water column, that then dissolved during the beginning of the incubation). This may have reduced dissolved Fe concentrations in the first sample of this incubation through oxidative removal. However, excluding this first value would not affect the overall flux magnitude derived from this incubation.

At stations below the permanent oxygen minimum zone (station 9 and 10) the oxygen was not consumed during the incubations, which is why it is unlikely that endobionts came to the sediment surface.

Referee comment 12:

4.1.2. I am not convinced about the half-life calculations since there is little change in iron concentrations and they do not fit with a reaction transport model to me. What about these data being colloidal fractions that pass the 0.2 μm poresize of the filter that would have little reactivity as shown by the slight slopes that look mostly within error bars? I agree that 1 point from station 8 and two from station 1 are clearly above error bar range but only station 8 seems to indicate that you caught the BBL. However, station 1 profile points to 2 homogeneous layers that suggest diffusion is not dominant there Silica data should appear somewhere

Response to referee comment 12:

The determination of the removal rates of Fe in near bottom waters are based on the assumption that silica is transported vertically by eddy diffusion and the modelled eddy diffusion coefficient from our data determines the half-life of Fe in the first 4 m above the seafloor. It is possible that our assumption of solute transport by eddy diffusion is not correct. Alternatively, decreasing Fe and Silica concentration above the seafloor could be due to super-imposed water layers with different Fe and silica concentrations but little vertical exchange. In this case our calculated half-life would be an underestimation. We are very aware of these limitations and will expand section 4.1.2 to explicitly discuss the uncertainties of this method.

We only applied the half-life calculations at stations with a discernable gradient within the first 4 m above the seafloor. This is clearly the case at station 4, but even though the gradient at station 3 falls within error we decided to also use this station because it still shows a clear gradient when compared to silica.

The silica concentrations measured in near bottom waters will be listed in a table in the appendix.

Referee comment 13:

4.2.1. Line 632 is it possible to detail how CdS precipitation in the near bottom water column was calculated? Show linear regressions, statistical significance and determination coefficients. What are the ranges in table 3? This section is very interesting but I feel uncomfortable with the lack of details about calculations and numbers out of it. It is a pity because it is I guess the most important part of the study

Response to referee comment 13:

The determination of the particulate delivery of Cd to the sediment via CdS precipitation in the water column is explained below table 3. We will also add an explanation to the main text in section 4.2.2 Quantification of the sedimentary Cd sink.

We will plot the linear regressions for the concentration change over time during the benthic chamber incubation for Fe and Cd (figures 5 and 7). We will also mention the coefficient of determination for each linear regression.

The ranges in table 3 listed in column “Cd from organic matter” result from the minimum and maximum estimate of Cd that is delivered to the sediment via organic material (see below table 3 and explanation in section 4.2.2). These values were determined by multiplying the average concentration ratio of Cd to C in phytoplankton (Moore et al., 2013) with published particulate organic carbon rain rates (maximum estimate) or burial rates (minimum estimate) for each individual site (Dale et al., 2015). The ranges in table 3 in the column “CdS precipitation in water column” result from the subtraction of the minimum and maximum of Cd that is delivered by organic matter, we add this information to the explanation below table 3.

Referee comment 14:

Conclusions Station 1 seems to weaken the first statement of the conclusion The order of TM affinity towards sulfide is shown for the first time in the conclusion. It should be said more explicitly in the introduction

Response to referee comment 14:

We will add to the introduction in line 60 the order of decreasing sulfide solubility for several mono-sulfide forming trace metals (e.g. Fe>Zn>Cd).

Referee comment 15:

The expansion of OMZ is evocated to draw a future scenario of TM burial/recycling within the OMZ but what about seasonality and other cyclic controls such as el nino/la nina conditions? Conclusions seem a bit too bold in that way. I am not sur that a putative enhancement or decrease of TM upward flux to surface waters (that is by the way not shown here with the Cd example) would affect marine ecology. At least nothing in the paper discusses it. If I read only the conclusion and the abstract, this will be the take-home message of the paper I would guess and the paper does not address such important question.

Response to referee comment 15:

In our manuscript we want to focus on long-term environmental changes, even though we are aware that seasonality or non-cyclic oceanographic and meteorological features as ENSO are important to consider. There are other publications dealing with these short term changes e.g. by Scholz et al., 2011, who discussed the impact of ENSO-related oxygen fluctuations on the early diagenesis of redox-sensitive trace metals.

The concentration/reservoir of a trace metal in the ocean is dependent on the respective input and output of this element. Consequently, benthic recycling and burial processes are an important aspect of oceanic mass balance. As especially Fe and also other trace metals can be often the limiting factor for primary productivity the availability of TMs has a direct impact on phytoplankton growth. As bottom water can be upwelled to the surface ocean, the benthic burial or release of a trace metal can have an impact on the amplitude or fuel/limit primary productivity. We will refer to this last statement in the introduction.

Technical referee comment 1:

Line 189 “compared” Line 191 “covered” Line 207 “weighted” Line 317 what are optopodes? Line 563 “can take”

Response to technical referee comment 1:

These mistakes will be corrected.

Technical referee comment 2:

Figures 5 and 7 should show linear regressions and equations as well as error bars for the data points. Then, equations and quality of regressions should be quickly described to prepare the discussion section about iron being oxidized by nitrogen compounds

Response to technical referee comment 2:

We will implement these suggestions. We will plot linear regressions for the concentration change over time during the benthic chamber incubation for Fe and Cd (figures 5 and 7). We will also mention the coefficient of determination for each linear regression, which will also be briefly described.

Technical referee comment 3:

Details from saturation indices shown in the caption of figure 10 should maybe appear within the text of the manuscript in the methods or the discussion

Response to technical referee comment 3:

We list the order of sulfide solubility for different trace metals in the discussion in lines 687 - 689.

Technical referee comment 4:

Table 3 what is within brackets for station 1 flux from chambers. This calculation would show what evolution of Cd concentration within the chamber? Is that coherent with your data?

Response to technical referee comment 4:

In table 3 the numbers in brackets represent the flux calculated from the Cd concentration difference between the bottom water (0.5 m) and the first sample from the benthic chamber (taken after 0.25 h) which was characterized by near-complete Cd depletion. This is explained below table 3.

The flux is coherent with our data in the sense that it would be high enough to explain the Cd enrichment in the sediment. We will explain that in section 4.2.2 Quantification of the sedimentary cadmium sink.

Technical referee comment 5:

Figure 9 reminds me a figure from my paper about cadmium fluxes in a seasonal hypoxic lagoon (Metzger et al., 2007) with a kind of threshold. You could put in the x-axis oxygen concentration and or sulfide and or nitrate. Why did you do only a figure for iron?

Response to technical referee comment 5:

In figure 9 the spatial extension of biogeochemical conditions are indicated by bars above the figure (oxygenated, nitrogenous, sulfidic).

Indeed, it would be very interesting to compare also the variability of Cd fluxes between different seasons or years under changing biogeochemical conditions. Unfortunately, however, no benthic Cd fluxes were determined or published prior to our 2017 cruise.

References

Dale, A. W., Sommer, S., Lomnitz, U., Montes, I., Treude, T., Liebetrau, V., Gier, J., Hensen, C., Dengler, M., Stolpovsky, K., Bryant, L. D. and Wallmann, K.: Organic carbon production, mineralisation and preservation on the Peruvian margin, *Biogeosciences*, 12(5), 1537–1559, doi:10.5194/bg-12-1537-2015, 2015.

Lenstra, W. K., Hermans, M., Séguret, M. J. M., Witbaard, R., Behrends, T., Dijkstra, N., van Helmond, N. A. G. M., Kraal, P., Laan, P., Rijkenberg, M. J. A., Severmann, S., Teacă, A. and Slomp, C. P.: The shelf-to-basin iron shuttle in the Black Sea revisited, *Chem. Geol.*, 511(April), 314–341, doi:10.1016/j.chemgeo.2018.10.024, 2019.

Metzger, E., Simonucci, C., Viollier, E., Sarazin, G., Prévot, F., Elbaz-Poulichet, F., Seidel, J.-L. and Jézéquel, D.: Influence of diagenetic processes in Thau lagoon on cadmium behavior and benthic fluxes, *Estuar. Coast. Shelf Sci.*, 72(3), 497–510, doi:10.1016/j.ecss.2006.11.016, 2007.

Moore, C. M., Mills, M. M., Arrigo, K. R., Berman-Frank, I., Bopp, L., Boyd, P. W., Galbraith, E. D., Geider, R. J., Guieu, C., Jaccard, S. L., Jickells, T. D., La Roche, J., Lenton, T. M., Mahowald, N. M., Marañón, E., Marinov, I., Moore, J. K., Nakatsuka, T., Oschlies, A., Saito, M. A., Thingstad, T. F., Tsuda, A. and Ulloa, O.: Processes and patterns of oceanic nutrient limitation, *Nat. Geosci.*, 6(9), 701–710, doi:10.1038/ngeo1765, 2013.

Noffke, A., Hensen, C., Sommer, S., Scholz, F., Bohlen, L., Mosch, T., Graco, M. and Wallmann, K.: Benthic iron and phosphorus fluxes across the Peruvian oxygen minimum zone, *Limnol. Oceanogr.*, 57(3), 851–867, doi:10.4319/lo.2012.57.3.0851, 2012.

Pakhomova, S. V., Hall, P. O. J., Kononets, M. Y., Rozanov, A. G., Tengberg, A. and Vershinin, A. V.: Fluxes of iron and manganese across the sediment–water interface under various redox conditions, *Mar. Chem.*, 107(3), 319–331, doi:10.1016/j.marchem.2007.06.001, 2007.

Point, D., Monperrus, M., Tessier, E., Amouroux, D., Chauvaud, L., Thouzeau, G., Jean, F., Amice, E., Grall, J., Leynaert, A., Clavier, J. and Donard, O. F. X.: Biological control of trace metal and organometal benthic fluxes in a eutrophic lagoon (Thau Lagoon, Mediterranean Sea, France), *Estuar. Coast. Shelf Sci.*, 72(3), 457–471, doi:10.1016/j.ecss.2006.11.013, 2007.

Rapp, I., Schlosser, C., Rusiecka, D., Gledhill, M. and Achterberg, E. P.: Automated preconcentration of Fe, Zn, Cu, Ni, Cd, Pb, Co, and Mn in seawater with analysis using high-resolution sector field inductively-coupled plasma mass spectrometry, *Anal. Chim. Acta*, 976, 1–13, doi:10.1016/j.aca.2017.05.008, 2017.

Scholz, F., Hensen, C., Noffke, A., Rohde, A., Liebetrau, V. and Wallmann, K.: Early diagenesis of redox-sensitive trace metals in the Peru upwelling area - response to ENSO-related oxygen fluctuations in the water column, *Geochim. Cosmochim. Acta*, 75(22), 7257–7276, doi:10.1016/j.gca.2011.08.007, 2011.

Scholz, F., Schmidt, M., Hensen, C., Eroglu, S., Geilert, S., Gutjahr, M. and Liebetrau, V.: Shelf-to-basin iron shuttle in the Guaymas Basin, Gulf of California,

Geochim. Cosmochim. Acta, 261, 76–92, doi:10.1016/j.gca.2019.07.006, 2019.

Shibamoto, Y. and Harada, K.: Silicon flux and distribution of biogenic silica in deep-sea sediments in the western North Pacific Ocean, Deep Sea Res. Part I Oceanogr. Res. Pap., 57(2), 163–174, doi:10.1016/j.dsr.2009.10.009, 2010.

Sundby, B., Anderson, L. G., Hall, P. O. J., Iverfeldt, Å., van der Loeff, M. M. R. and Westerlund, S. F. G.: The effect of oxygen on release and uptake of cobalt, manganese, iron and phosphate at the sediment-water interface, Geochim. Cosmochim. Acta, 50(6), 1281–1288, doi:10.1016/0016-7037(86)90411-4, 1986.

Turetta, C., Capodaglio, G., Cairns, W., Rabar, S. and Cescon, P.: Benthic fluxes of trace metals in the lagoon of Venice, Microchem. J., 79(1–2), 149–158, doi:10.1016/j.microc.2004.06.003, 2005.

Warnken, K. W., Gill, G. A., Griffin, L. L. and Santschi, P. H.: Sediment-water exchange of mn, fe, ni and zn in galveston bay, texas, Mar. Chem., 73(3–4), 215–231, doi:10.1016/S0304-4203(00)00108-0, 2001.