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Interactive comment on "Thermal and haline effects on the calculation of air-sea CO$_2$ fluxes revisited" *by* D. K. Woolf et al.

D. K. Woolf et al.

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1 General Comments This article is concerned primarily with the issue of the thermal skin effect on the airsea exchange of carbon dioxide. There is also discussion on the effect of diurnal warm layers, as well as the impact of a salinity skin on the air-sea fluxes of CO2.

These are significant contributions of our paper, but we note that we have also outlined an approach to deal with variability in the upper ocean on small time scales and spatial scales in the context of climatological calculations.

This article criticizes the paper from McGillis and Wanninkhof (2006), who showed that the differences in the thicknesses of the thermal and mass boundary layers had

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not been properly considered in previous results, and this led to an over-estimate of thermal effects on gas fluxes.

We presume the reviewer chooses his words advisedly and thinks that McGillis and Wanninkhof "showed .... an overestimate of thermal effects". The paper of McGillis and Wanninkhof has many merits but we have shown that their specific conclusion on thermal skin effects is erroneous. The reviewer has not provided any argument that refutes our finding.

This article argues that the existence of the cool-skin temperature deviation can in fact influence the oceanic uptake of CO2 by up to 0.6 GtCyr-1.

This statement is correct, but rather misleading. We provide a simple calculation of the thermal skin effect that suggests the value will be in the range 0.1-0.6 Pg C yr -1.

This paper was long and difficult to read and I think it has several issues.

We acknowledge that the paper is not an "easy read" and it is our responsibility to make it as readable as practical, but there are some impediments. The subject is important and requires a fairly arcane combination of boundary layer physics and carbonate chemistry to untangle. Also, given that some previous accounts contain errors as a result of missing specific details, we think a rather pedantic treatment of some key issues is necessary. Rather than identifying any extraneous material, the reviewer seems to require some digressions that would make it longer and in our opinion would spoil the flow of the paper.

We are grateful for some specific points made by this reviewer and various comments by other reviewers that we will use to produce an improved revised version. We do need to work harder at the presentation and we are committed to this enterprise.

In general, there are several missing references, which would indicate that the authors are not familiar with the literature (or they are being too selective).

The reviewer tries to read more into our omission of a few particular references than we

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think is fair. Having said, we thank the referee for mentioning these references, which do all have merit, as we will discuss below as each occurs.

There are also some fundamental flaws with their argument which I have described below.

The perceived flaws will be dealt with individually below, but in summary we find that the reviewer has not identified substantive flaws in our argument. Instead, several of the comments imply that the reviewer has an insufficient understanding carbonate chemistry and boundary layer physics to grasp our meaning. Given that the reviewer will not be alone in this struggle, we will strengthen the description of the basic underpinning science.

2 Specific Comments Section 2 describes the three mechanisms mentioned in the introduction. It would make it easier to read if the authors were to provide a few sentences at the beginning of the section, and then describe each mechanism in subsections.

We think this is a good suggestion, thank you, and will adopt it.

The discussion around Figure 1 does not include the haline boundary layer, even though this is in the title of the article. The authors should include discussion of the salt boundary layer here. There are also several different temperatures being introduced, and it would be useful to have an explanation of these in the figure caption. Also, there are a lot of different temperatures, concentrations, etc. (e.g. TB, TM), and it would be useful if these could be summarized in figure 1 which could be used as a reference when reading the paper. It would be useful to know that the authors are at least aware of how XCO2 is determined in equation (1), and therefore it would be very useful to have a brief description of measurement techniques.

We agree that it will be sensible to mention salt early in section 2, but only briefly and we would delay detailed discussion until Section 7. As stated in Section 7, since the Schmidt numbers of salt and carbon dioxide are rather similar, the profiles of their

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boundary layers (mass boundary layers) will be very similar and unlike the case for heat, there is not a substantial decoupling of salt and gas. We will add a sentence to explain this. We plan to leave it at that, since it will be confusing to introduce more than one mass boundary layer into any of the figures. It was our intention that Figure 1 helps to summarise and introduce the definitions used later in the paper. We agree that this will be more effective if the caption is extended to summarise the definitions.

We are aware of how XCO2 is measured (or inferred, if moist air is measured) but describing those techniques in this paper would be an irrelevant digression. We can think of no legitimate reason for raising this issue.

Equation (2) introduces solubility and the authors describe how different solubility formulas introduce uncertainties. However, they do not indicate which formula they follow. Can the authors recommend one over the other? In figure 2 they present the formula by Weiss (1974) - please explain the arguments for accepting the Weiss formula? With respect to figure 2, I think the authors should plot the temperature sensitivities for salinities of 30-35 ppt, as I think (in general), there is less attention given to salinity than temperature.

We should have and can be more explicit that all dependent calculations described in the text are based on the formulae of Weiss. We are clear that these are preliminary calculations. Since we only quote results to 2 significant figures, while the values from Weiss (1974) and from Li and Tsui (1971) typically differ by 2% in warm water (the worst cases), the results barely depend on the choice of formulae, as long as that choice is consistently applied. We shall be clear that the selection does not represent an endorsement of Weiss (1974) over alternative formulae. At the top of P16387, we point out that this is a source of uncertainty. In all of this we follow McGillis and Wanninkhof (2006) who pointed out this significant source of uncertainty. We think their finding is worth repeating, but like them we cannot make a simple statement of which formula is best.

The sensitivity of solubility to temperature varies considerably across the natural temperature range, a fact that is often overlooked; therefore this dependence deserves pride of place. The variation of this sensitivity with salinity is relatively slight across the usual oceanic range of salinities. We don't have any great objection to including curves for other salinities in the figure. What it shows is not that interesting, but it does add some information. The sensitivity of solubility to salinity as a function of temperature is shown in Figure 6. In response to a comment of Anonymous Reviewer 2, we are considering whether Figures 2 and 6 should be combined.

The discussion regarding the calculation of CM: I agree that it is possible to determine CB with the measurements from a shipboard system. However, I cannot possibly understand how the authors propose to have a measurement of the temperature and salinity at the base of the mass boundary layer i.e. TM and SM. Can they describe how such a determination can be made? I also have a hard time accepting the assumption that the DIC and TA will remain unchanged between a depth of several meters and zM (about 100 μm). I would like to see a justification of this statement here (rather than later as they suggest).

We think that the structure of the paper is entirely logical in this case and would not benefit from (what would be in our opinion) a poorly-placed digression. We are clear in the Introduction that Section 2 identifies mechanisms and gives an introduction to their quantitative importance. Practical calculations are delayed to later sections, after other pertinent facts are introduced. In the particular case mentioned, the delay is only to the next section, which is already made clear.

We state an assumption across the page break of P16387 and P16388 that "we have available estimates for the temperature and salinity {at "M"}"; we do not state or imply any direct measurements at "M". The reviewer should be aware (since a later comment is directed to this point) that later sections describe inference of the temperature (and in section 7, salinity) at "M". We find the comments regarding DIC and TA surprising, since the assumption that these are conserved quantities in particular situations

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is a widely applied and often an indispensable aid in applying the theory of carbonate chemistry to practical problems. In fact, we have been relatively pedantic in this paper in explicitly stating the assumption and we have been careful to limit its application to where it can be justified, rather than an implicit assumption as in many other studies. The paper already contains an explanation (P 16388, lines 4-20). In the following paragraphs, we reinforce the underpinning reasoning (we think this full description would be an unwelcome digression in the paper itself, but we will make sure the description of carbonate chemistry is cogent in the revised paper).

Firstly, we summarise some pertinent details of the theory of carbonate chemistry, but refer the reviewer or reader to a good text book for a more thorough account (see for example, Chapter 6 of Williams and Follows, "Ocean Dynamics and the Carbon Cycle", Cambridge University Press, 2011; or Chapter 4 of Emerson and Hedges, "Chemical Oceanography and the Marine Carbon Cycle", Cambridge University Press, 2008). While other carbonate parameters (including fugacity) are sensitive to temperature, DIC and TA are unique in being conserved for a change in temperature or pressure. In simple terms, conservation of DIC represents mass conservation, while conservation of TA represents a conservation of charge. One approach for modelling the carbonate system in the ocean (see for example: Brostrom et al., 2000, Marine Chemistry, 72, 151-169; Kuss et al., 2004, Tellus, 56B, 175-182; for attempts to close annual or seasonal budgets of carbon) is to assume either conservation or a calculable change in DIC and/or alkalinity. Neither DIC nor Alkalinity is conserved by every process. Williams and Follows (see Figure 6.16) identify "organic matter formation", "calcification" and "dilution" as processes that change both DIC and Alkalinity, while the air-sea exchange of carbon dioxide breaks mass conservation (DIC) but does not affect charge balance (TA). Working in terms of DIC and TA is a sensible approach where fugacity must be expected to vary in response to temperature changes.

Secondly (to explain the precedent to our approach), we note that DIC and TA conservation within a vertical profile is either explicit or implicit in all studies of upper ocean

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carbon dioxide and air-sea exchange that we can find. In most cases this takes the form of assuming a sensitivity of fugacity to temperature of 4.23% / K since this is based on an empirical measurement of the change in fugacity "at constant chemistry" (Takahashi et al., 1993). Thus, whenever "4.23% / K" is invoked there is an implicit assumption that DIC and TA are conserved. The application of either "4.23%" or the more accurate temperature dependent form is explained by Takahashi et al. (2009; Sections 2.2 and 6.4.4). As discussed in the paper, McGillis and Wanninkhof (2006) explicitly discuss DIC and TA conservation and suggest a more sophisticated approach than Takahashi, but the results are broadly similar.

Finally, for the specific case of profiles between "B" and "M", we can cite the precedent in calculations for warm layers e.g. Olsen et al., 2004). Specifically, Olsen et al. (2004, GRL, doi:10.1029/2004GL020583) apply 4.23% to calculate the daytime and nighttime anomalies of partial pressure used in their calculations; an assumption of constant DIC and TA is implicit. In most situations (i.e. excluding "warm layers" or surface pools formed by rainfall), the assumption of a constant DIC and TA between "B" and "M" is trivial since this part of the water column will then be "well mixed" and both DIC and TA are invariant with respect to pressure change.

In summary, we are doing something very conventional and robust in assuming that DIC and TA are conserved within the vertical profile.

Please add the reference Wanninkhof and Thoning (1993) to P16388 L25 (i.e. correct shipbased measurements using an equilibrator).

Our description of equilibrator-based measurements and associated thermal corrections is largely taken directly from Pfeil et al. (2012) and is appropriately referenced. The procedures for equilibrator-based measurements have been honed over the last two decades and have been explained in several papers and manuals. Having said all that, Wanninkhof and Thoning (1993, Marine Chemistry, 44, 189-204) were pioneers and the modern methods are only a slight refinement of the systems that they describe

and we can easily incorporate that reference.

The authors state that the temperature of interest for measurements is the "climatological" temperature at the base of the MBL. They also state that if there is a discrepancy between the measured temperature and the "temperature of interest" (arbitrarily a climatological one), then "the same equation can be applied". I don't understand this argument or its relevance. Can the authors kindly explain? Also, once again, the authors postpone a discussion (P16389 L1) which only adds to the confusion.

Again the reviewer wishes us to interrupt the flow of the paper with a digression and again we think that this is a poor suggestion. Again, we are applying an iso-chemical temperature dependence just as in numerous previous studies. As promised, the point is fully explained in Section 3 (we will clarify this location) and any reader should be able to wait until the appropriate section.

The sentence "but importantly any error in the in situ data is otherwise irrelevant to the calculation" does not make any sense. If there are errors, then surely these would be relevant? Am I missing something?

This quote is taken out of context (from line 7-8 of P16389) and only sounds a little odd in isolation. The text from line 21 of P16388 to line 13 of P16389 explains the situation fully. The key here is a distinction between the direct measurements within the hull of the ship, "the equilibrator measurements", and data inferred for outside the hull of the ship, the so-called "in situ data". We explain that for climatological purposes the equilibrator data is more useful and the inferred in situ data can be largely discarded.

Figure 3 is a "common example" of vertical temperature structure. Can the authors provide some references? It's also described as a "common but minority situation", which doesn't make sense. I'd like to see the following statements backed up with some citations: - Therefore, some sea surface temperature climatologies use only nighttime data to calculate a foundation temperature. - Since changes in total DIC and alkalinity within one day are usually miniscule it is usually reasonable to assume that these

properties are uniform within this layer. – A very thin layer at the sea surface largely controls gas transfer due to the suppression of turbulence in that layer.

We discuss warm layers in Section 6 and provide several references on the subject. We think our statement is uncontroversial. As noted by the other Anonymous Reviewer, the thermal profile of the oceans is well known to physical oceanographers. Warm layers are discussed in many physical oceanography text books and we are puzzled by the reviewer's query. Many detailed accounts our available in the research literature. There is no harm in citing more of these, but it seems to us to be reasonable to delay a referenced description until Section 6.

There is no contradiction between "common" and "minority". The latter simply implies occurrence of < 50% and of course 10% or even 1% might be considered "common" rather than "rare". As mentioned in Section 6 on warm layers the frequency of warm layers varies from region-to-region (Stuart-Menteth et al., 2003) and thus the occurrence of such features is difficult to summarise in a few words, and "common but minority" remains a fair summary.

The remaining comments again concern referencing and the reviewer prefers us to supply documentation for some statements.

The use of daytime or nighttime measurements is generally buried within "user manuals" for particular data sets. GHRSST-PP (http://ghrsst-pp.metoffice.com/pages/sst_definitions/index.htm) defines foundation temperature more rigorously than our usage, such that both daytime and nighttime measurements should be used, but some quick "browsing" reveals that for sensible pragmatic reasons, some data sets present "foundation temperatures" that are based only on nighttime measurements (e.g. http://gosweb.artov.isac.cnr.it/index.php?option=com_content&view=article&id=81).

The query about changes in DIC and TA recalls the earlier comment on assuming

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these are constant within a vertical profile. Here the issue is whether it is reasonable to assume they are conserved through daylight in a warm layer, or for a full day generally in a mixed layer. Again, we can recall that this assumption is implicit in previous warm layer studies (e.g., Olsen et al., 2004, GRL, doi:10.1029/2004GL020583), but we do agree that the issue deserves a little more comment and referencing in this instance. For instance, we should note that McGillis and Wanninkhof (2006, Figure 2 and associated text) recall observations from GasEx 2001 previously reported by McGillis et al. (2004, J Geophys Res, C08S02) where an excursion of DIC implies a significant effect on fugacity. More generally, this calculation depends on estimate of net productivity and other processes. We will revise the text slightly, but don't want to digress too far. A general reference to the slow seasonal variation in the carbonate system (e.g. Brostrom et al., 2000, Marine Chemistry, 72, 151-169) seems most appropriate.

Figure 4 is not drawn to scale (P16395 L9), but should be. The depth axis can be drawn with a log scale. I think it's important to show the different scales we are dealing with for temperature and mass. Do we know that the temperature profile with the thermal boundary layer has the shape which is shown in Figure 4? Can the authors provide a reference for this? Also the caption in figure 4 should indicate what each of the various temperatures and concentrations represent (see earlier comment). I would like to see lengthscales added to Figure 4, which will require an estimate of the various boundary layers (for estimates of the thermal boundary layer thickness, see Ward, Air–Water Interfacial Temperature Measurements in "C.S. Garbe, R.A. Handler, B. Jähne (eds.): Transport at the Air-Sea Interface pp. 191-201, 2007, Springer-Verlag Berlin, Heidelberg 2007").

We will add to the caption to remind the reader of the identity of "I" "Rad", "M", "T" and "B". We think the request that Figure 4 is drawn to scale is misplaced. A scale drawing is appropriate where a very specific boundary layer model is discussed, but here it is self-evident that a schematic representation is much more helpful. The substantive requirement is that the different scales of the thermal and mass boundary layers are

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evident and that is clearly achieved in the figures and the text. A (semi-)logarithmic plot is a bad idea since the sense of the "curvature" which is clear in the schematic would be obscured by a logarithmic vertical scale (also the interface disappears off scale!). Yes, we do know that the mass and thermal profiles in the boundary layer must be curved as shown (schematically!), though there is plenty of room for debate on a more quantitative description. The precise curvature and dimensions of each layer depend on the specific boundary layer model. We suggest referring to Fairall et al. (2000, Boundary-Layer Meteorology, 96, 63-105) for a good example of a "bulk layer model" (though noting that the actual curvature is distorted in their Figure 2 since they use a semi-logarithmic scale!) and two papers by Asher, Atmane and Jessup (Atmane et al., 2004, J Geophys, Res, 109, C08S14; Asher et al. 2004, J Geophys, Res, 109, C08S12) for an alternative description in terms of penetration theory. All of these theories demand a curvature in the profiles for both concentration and temperature AND require turbulence within the viscous boundary layer.

{here we note that the above comment and response lead naturally into a later comment and response, therefore we have moved that comment forward to here}

Further down they state that "below the base of the mass boundary layer, turbulence will be substantial and therefore transport faster than by molecular diffusion alone". This is thoroughly inaccurate. There is no turbulence available for mixing at this depth, as it is embedded within the viscous sub-layer where turbulent diffusion is damped, and where molecular viscosity is the dominating momentum transport mechanism. However, the rate of strain of the concentration fields results in enhanced mixing rates of gases by molecular diffusion. The authors do not seem to understand the issues here.

The reviewer is entirely wrong. Here and in the earlier comment on the profile of temperature and concentration in the boundary layers the reviewer seems to be influenced by a simplistic model of boundary layer physics that has been thoroughly discredited. Again, we can refer to Fairall et al. (2000, Boundary-Layer Meteorology, 96, 63-105) and Atmane et al. (2004, J Geophys, Res, 109, C08S14) for descriptions that are cred-

ible, support our description and thoroughly refute the statements of the reviewer. As summarised in the first paragraph of Section 4 of our paper, one of the earliest models of air-sea gas transfer, "the stagnant layer model" envisaged a turbulence-free layer of typically 30-35 μm thickness, with fully turbulent ("well mixed") conditions below. A significantly thicker stagnant layer would simply be inconsistent with known rates of air-sea gas transfer. This and similar models for heat and momentum can be used to calculate an appropriate scale of the sub-layers for each of gas, temperature and momentum. However, as explained in Section 4, these scale lengths remain a useful abstraction, but they should not be taken literally. Fairly thick stagnant or laminar layers are simply inconsistent with our knowledge of gas, heat and momentum transfer immediately below the sea surface. More successful models either specify that the influence of turbulence reduces gradually to the very interface (Fairall et al., 2000, see especially Figure 3 where the vertical profile of "total diffusion coefficient" is plotted), or that there is only a very thin layer free of turbulent eddies (see for example Table 4 of Atmane et al. 2004, where thicknesses, "approach distances" in penetration theory, of 6-40 μm are inferred for various laboratory experiments). In summary, the paradigm of a thick laminar sub-layer with no turbulence is discredited for fluid motions below the sea surface, and instead the description given in the paper is readily substantiated. We propose to add references to Fairall et al. (2000) and Atmane et al. (2004), since these will serve to explain that a precise description of the vertical profiles remains open to debate.

For the discussion of TRad, the article by Donlon et al. (2002) should be cited ("Toward Improved Validation of Satellite Sea Surface Skin Temperature Measurements for Climate Research"). This Donlon paper also introduces the nomenclature SSTskin, so I don't understand why the authors chose to omit this from their list of references.

To the first author's direct knowledge, terms like radiometric temperature, skin temperature and sub-skin temperature were widely discussed in the community in the 1990s, before several being adopted as standard terms through GHRSST. We did not use the

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proposed reference since we were not concerned with validation, but the reviewer is certainly correct that the paper gives a cogent description of these terms, which makes it a useful reference that we should have used and will use in a revised version.

The authors make the point that TRad should not be used as a substitute for TM, but instead TB should be used. This is not correct. It is often the case that TM « TB (where TB is measured at several meters below the surface) e.g. under conditions of diurnal warming. The authors should refer to Ward (2006, JGR, doi:10.1029/2004JC002689). The authors go on to discuss the concentration profile in Figure 4, and state that the water below the depth M is well-mixed.

The reviewer is aware that we wrote an entire section on warm layers, since this was noted in the first sentence of the review. However, the reviewer has chosen to take one or more (unidentified) individual phrases out of context. Even then, the reviewer has misquoted those phrases. We have been very careful to distinguish between cases where it is reasonable to assume that an available bulk temperature is a pragmatic substitute for the temperature at the base of the thermal skin (at "T", not at "M" as stated by the reviewer), and "warm layer" cases where that is not a sensible assumption. We state that in the absence of a warm layer, the bulk temperature will often be the best available substitute for an actual measured temperature at "T". This contention is shared with numerous previous studies and indeed is consistent with Donlon et al. (2002) who state (pages 355-356) : "... SSTsubskin is typically replaced by SSTdepth. ...Clearly it is important to recognizes that (1b) now includes both the skin temperature deviation and the effect of thermal stratification that may be present in the upper ocean ...". Thus, we have included Section 6 to cover cases where there may be thermal stratification such that temperature at "T" may be significantly warmer than the bulk temperature.

The nomenclature SSTsubskin is used further in the paper, but the source of this term is not cited - Donlon et al. (2002).

Again, the term has been used since the 1990s and our adoption of it was independent of that specific paper, but we are happy to include the reference suggested.

Equation (6) is simplified to equation (7) by assuming that CI = CM. The same is assumed in equation (12). If I look at Figure 4, I see a concentration (rapid) profile where the only variability that exists with depth is the gradient between the depths I and M. But here the authors now assume that there is no gradient. Does this not eliminate their whole argument?

On the contrary, equation (7) (and similarly equation (13) for warm layers) simply describes a useful approximation for "near-saturation conditions" (CI $\sim$ CM). There is absolutely no implication that there is never a gradient. A net flux requires that gradient.

Add a reference by Ward et al. (2006, JGR, doi:10.1029/2003JC001800) at P.16400 L.24 (observations of warm layers).

Section 6 on warm layers was adequately referenced but we have no particular objection to adding this reference, which displays some practical examples.

I'd like to see a diagram showing the warm layer situation described on P16401.

A warm layer situation is depicted in Figure 4

There is no reference to Olsen et al. (2004, GRL, doi:10.1029/2004GL020583) when discussing the effect of the warm layer on gas transfer.

As noted earlier this is a useful example of a warm layer study that we are happy to mention. This particular study was based on warm layer characteristics derived by Stuart-Menteth and colleagues, who we had already cited (Stuart-Menteth et al., 2003, J Geophys Res, 108, 3155) and is similar in approach to Kettle et al. (ACP, 9, 529-541), which we had also cited, therefore its inclusion will not alter any conclusions.

There is no reference to Turk et al. (2010, GRL, doi:10.1029/2010GL045520) when

discussing rain on P16405 L27.

We are familiar with this paper, which exploits previous work in a modelling framework. It is a relevant paper and we have no objection to including it, though it should not be necessary.

The conclusions section has the sentence: "Where a deep or nighttime temperature is applied when a warm layer is in place, the interfacial concentration will be overestimated and the concentration at the base of the MBL will be underestimated". I don't get this, and I would need to see a diagram for this to be adequately explained. Please provide such a diagram.

The first thing to note is that this sentence is taken from the Conclusions and nobody should expect a lengthy explanation here, but should refer to the detailed section from which this conclusion is drawn. Here, it should be readily apparent that this conclusion is drawn from Section 6 on "warm layers". The conclusion that ". . .the interfacial concentration is overestimated" and thus corrected down is expressed by equation (10) and explained in the preceding paragraph. The conclusion that " the concentration at the base of the MBL will be summarised" is similarly expressed by equation (11) and explained in the preceding paragraph. We do not think adding a diagram will be beneficial.

3 Technical Corrections P16383, L25: it would be useful if the authors could list the three mechanisms here in the introduction, so that the reader knows what to expect.

We prefer the reviewer's earlier suggestion that the three mechanisms are introduced at the start of Section 2.

P16385, L5: remove "or Molecular Boundary Layer" and please refer to this layer as the mass boundary layer

We don't agree. We also prefer the term "mass boundary layer" and use it thereafter. However "molecular boundary layer" is also used in the literature and by mentioning

the alternative term once only, we should avoid any potential misunderstanding.

P16385, L13: delete "of the volatile"

Why? The sentence is technically and grammatically correct.

P16386, L9: an sensitivity should be a sensitivity

This will be corrected

P16387 L19: A new paragraph at "We now consider"

We agree that this would be an improvement and propose to put a sub-section break at this point.

P16395 L6: The following text is very unclear and should be re-written: "will be curved as the hand over to turbulent transport progresses further as the interface recedes"

We think that this should be clear enough except to a reader who has an erroneous vision of the structure of boundary layers (see detailed responses to the reviewer's comments on concentration, temperature and turbulence profiles). However, we will try to further clarify the description.

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