Response to reviews for the paper "Reconciliation of essential process parameters for an enhanced predictability of Arctic stratospheric ozone loss and its climate interactions"

We thank Susan Solomon, Jean-Pierre Pommereau, Rob MacKenzie and Ross Salawitch for detailed and constructive reviews that will help to improve our RECONCILE overview paper considerably. Below, we will comment on the more general issues that were raised more or less in each of the four reviews, and that will lead to a substantial reorganization of the paper. We will then respond in detail to specific points raised by individual reviewers.

A General response to all reviewers

(1) We will significantly reduce references to institutions and funding bodies in the main text.

(2) In the revised manuscript, we will identify more clearly the reconciliation that has been achieved. Then, the project title will hopefully not seem so inappropriate anymore. Obviously, there are important issues in terms of the predictive capabilities of polar ozone loss and climate interactions that have not been addressed by the project, and some of these issues are described in the outlook at the end of the conclusions. If the reviewers and the editor feel that the current title still does not fit the revised manuscript, we will change it to "Overview of activities and important results of the project RECONCILE".

(3) We will considerably shorten the "introductory part", sharpen the main outcomes, and elaborate on the results in terms of global modelling (in line with improving that part in the paper, cf. below).

(4) We will shorten the introduction (currently Sections 1 and 2) to eliminate the review character. The revised version will only mention the "standard information" found in most papers on polar stratospheric ozone loss, and then refer the reader to existing review articles. We will then focus on the motivation that led us to formulate the objectives of the project RECONCILE, using the questions posed in Section 2 of Jean Pierre Pommereau's review as a guideline. Following this path, i.e. opting for the "short route" rather than attempting to make our "historical background" complete, we will not cite some of the papers that were identified by the reviewers as "missing" in that Section. Nevertheless, we do agree that most of the suggested references should be cited in a comprehensive review of polar stratospheric ozone loss.

(5) The current Section 3 lists the RECONCILE strategy and activities, and we thank Jean Pierre Pommereau for supporting our viewpoint that this is needed. Nevertheless, we plan to shorten this Section. For the "standard activities" such as models, satellite products, etc., we will only state what the activity was used for in RECONCILE plus one or two references. Following the suggestion made by Rob MacKenzie, we will highlight the innovative tools and activities that moved forward the state of the art (e.g. the new $ClOOCl/Cl_2$ lab experiment, comprehensive PSC payload, the self-match flight strategy) and thus strengthen the "new science content" of this section.

(6) Sections 4 to 6 will be combined into one results sections, with headings referring to the RECONCILE key results rather than suggesting that we have addressed dynamics, microphysics and chemistry all in a fully comprehensive way. As in the Section on methods and activities, we will highlight the progress that has been gained during RECONCILE, referring to other previous and contemporary work wherever appropriate. We will place a particular emphasis on sharpening the Section on improvement of CCM simulations and implications for prognostic capabilities. Criticism raised by the reviewers on specific results or issues described in this Section will be taken into account. Here, this will be discussed in the response to points raised by individual reviewers below.

(7) We will include a more elaborate summary, highlighting where "reconciliation" has been achieved, and commenting on the implications of our results, in particular with respect to understanding and forecasting future Arctic ozone loss. With respect to our "outlook", we feel that after summarizing the results

of a large international research project such as RECONCILE, the identification of further research needs in the field by the large international team involved is not inappropriate for a scientific paper, and at least two reviewers seem to agree with that.

B Specific replies to Susan Solomon

Numbering refers to the numbering in the review. Some issues are being addressed by the general response above.

11) Some information on ozone climate interactions will be retained in the modified introduction, because it is one of the main foci of RECONCILE. But we will ensure a balanced representation of the state of the art and focus on the aspects relevant to the RECONCILE project.

12) We do not think that our sentence "A particular susceptibility of the Arctic stratosphere to climate change may manifest in the trend of cold stratospheric winters in the Arctic getting colder, amplifying heterogeneous chlorine activation and ozone depletion" suggest that the cooling trend is caused by climate change. Rather it is meant to emphasize that the Arctic ozone layer is particularly sensitive to any potential change in climate. But we will reword the manuscript to avoid any misunderstandings.

14) The English word “palpable” is used here with the exact meaning described in English dictionaries (both BE and AE). We do agree that the list of “areas of uncertainty” that we give in the first paragraph in Section 2.4 is too broad and may be interpreted as identifying uncertainties where there aren’t any. We will rephrase this introductory paragraph and then give concrete open questions that existed at the start of RECONCILE and represented the motivation for the project. We will include the stratospheric Br_y content as one of the parameters governing the rate of catalytic ozone loss.

15) Using a detailed comparison of CLaMS simulations to HAGAR high resolution tracer measurements, RECONCILE has shown that to correctly represent observed tracer fields and ozone loss in late winter, a correct initialization is critical, in particular with respect to the timing of vortex formation. If this is done correctly, then the existing state-of-the-art mixing parameterization does a reasonable job. We will change the rather general paragraph in the introduction to point more specifically into this direction.

16) RECONCILE has not resolved (and cannot resolve) uncertainties in reanalysis temperatures, and has not done any substantial investigation on the role of mountain waves in the Arctic vortex. However, these issues were not identified as “substantial uncertainties in polar ozone loss” (even though temperature biases can have a significant impact on atmospheric models) but were named as an explanation why previous observational studies on heterogeneous NAT nucleation were not conclusive. This has changed in the RECONCILE winter, where widespread CALIPSO observations of NAT without prior ice formation were made (described in Section 4.2).

17) As suggested, we will reference earlier work including Hanson et al., 1994.

18) Both, Wegner et al., 2012, and Wohltmann et al., 2012, address this issue and both try to retrieve more quantitative information on how large the impact of enhanced heterogeneous reactivity (on PSCs compared to cold binary aerosol) is on chlorine activation and ozone loss. Moreover, for a case in March 2005, Wegener et al., 2012 could analyse how much chlorine activation occurs on binary aerosols as opposed to PSCs (see comment 26 below). We wanted to present reasons why we deem this investigation important (as we all know, in the polar vortex, the temperatures at which PSCs form, and those, at which activation on cold binary aerosols becomes effective, are very similar, so the impact on polar ozone in models will be small). We do acknowledge that the statement “Clearly … on a global scale” is misleading in this context and will remove it.

22) The RECONCILE aircraft campaign has provided a set of high resolution measurements in the stratosphere, which were successfully reproduced with the mixing parameterisation currently implemented in

ClaMS. Insofar our "understanding of mixing" has not substantially advanced, but it has been demonstrated that it is possible to reproduce the rather complex fine scale structures in the polar stratosphere, subject to a correct initialization of the polar vortex (cf. comment 15 above) This will be discussed in the results section. The paragraph on existing CLaMS parameterisations in the "methods" Section will be shortened.

23) The sentence "Jackson and Orsolini's results showed that the data assimilation method is very promising and can lead to potentially more accurate ozone loss estimates than other, more established methods" on page 30681 will be removed. In the revised manuscript, we will draw sections 3.5 and 4.1.3 together and present a balanced discussion on the potential of the data assimilation method, and on shortcomings that need to be overcome.

24) The results on PSC microphysics are summarized in the abstract, and we will include a summary and implications (including the new PSC scheme for CCM, cf. below) in the new conclusions. The four key "in preparation" papers cited in this section have all been fully drafted now and we can sharpen this part of the results section even more.

25) The wording is indeed ambiguous here and will be modified. Nevertheless, we would like to point out that this introductory paragraph to Section 4.3.1. refers to cold temperatures and PSCs as "key to chlorine activation", and not "key to ozone loss".

26) a. In our opinion, Section 4.3.1 of this paper does not make the point that "binaries drive polar ozone loss". The same is true for Wegner et al., 2012. We will revise the text of the manuscript to make sure that this misunderstanding cannot occur anymore.

As we all know, polar ozone loss is driven by the complex interplay of a number of dynamic, chemical and microphysical processes. The frame and key components of that puzzle were first laid out by Susan Solomon in her 1986 paper. A central variable in almost all of the processes at play is temperature. Therefore, it may be justified to simplify into saying "low temperatures drive ozone loss" because they

- favour a stable and well isolated polar vortex
- lead to faster chlorine activation, either on PSCs or binaries
- favour PSC formation that leads to denitrification
- increase the rate of the ClO dimer cycle

This is the reason why ozone loss tends to correlate with [area] x [time below any reasonably low threshold]. While such temperature thresholds may be useful to make certain analyses or projections, they carry little to no information about the actual processes going on. Because RECONCILE aims at improving the mechanistic understanding of the processes relevant for polar ozone depletion and incorporating them in physical models, we have deliberately avoided a reference and discussion of such temperature thresholds.

Nevertheless, heterogeneous chlorine activation is a key step in the chain of processes leading to polar ozone depletion. While under most conditions it may not make a difference in models whether this occurs on PSCs or cold binaries, we still find it useful to elucidate the roles of different particle regimes for chlorine activation. This is notwithstanding the crucial role that PSCs play for denitrification and thus the length of the ozone loss season, as is discussed in much detail in Section 4.2.4.

b. We do not dispute the notion that chlorine activation and deactivation compete, and that it is the net between the two that matters. If temperatures are sufficiently low and particles are present (and Wegner et al., 2012, and Wohltmann et al, 2012, have shown that at least for the actual activation process, it is not so critical whether the surface area density is enhanced by the presence of PSCs), the deactivation reaction can actually enhance activation by partitioning Cl into $ClONO_2$ as a reac-

tion partner for HCl. To achieve complete activation of inorganic chlorine to ClOx, deactivation into $ClONO_2$ is actually required since HCl is far more abundant than $ClONO_2$ when the vortex forms.

c. The key point of the Grooß et al. (2011) paper is that "a balance is maintained by gas phase production of both HCl and HOCl followed by heterogeneous reaction between these two compounds". The paper does not, however, discriminate between PSCs vs. binaries, even though at some places it refers to the "PSC threshold" (note that in other places it refers to "195 K as the threshold temperature for chlorine activation"). Parameterisations for activation on all particle types were included in the CLaMS runs performed for that paper, and we really cannot discern whether it is the binaries or the PSCs doing the activation job at low temperatures. There is one sentence in the context of sensitivity of the model to denitrification (Section 3.2 of Grooß et al., 2011) that could be interpreted in a way that the PSC surface area is critical: "Second, less HNO_3 causes smaller surfaces of solid and liquid PSCs, which slows down the heterogeneous chlorine activation reactions". However, the effect of the (in this case reduced) additional surface area due to PSCs vs. binaries was not quantified in the Grooß et al. study. This quantitative investigation was done by Wegner et al., 2012, who showed that the effect of decreased surface area on chlorine activation is small.

d. As stated above, the whole discussion is about chlorine activation and not about ozone loss, and this is true for Figure 16 as well. It displays the temperature dependence of the activation reaction, and the enhancements presented by different types of PSCs over background aerosol at different temperatures. Obviously, for net chlorine activation and thus ozone loss rates, the balance with deactivation is important, but the most important factor influencing deactivation, HNO_3 removal by denitrification, is discussed in another section.

e. Wegner et al., 2012 tried to validate the reanalysis temperatures during the March 2005 flight and the days prior to the flight with in-situ temperature observations. PSC calculations in this paper are actually performed with a -1.5 K offset applied to temperatures, as is explained in detail in this paper. Of course, to be certain whether PSCs were present or not observations along the trajectories are necessary. However, such measurements do not exist so we have to rely on reanalysis data. Given all the uncertainties concerning PSCs, we wanted to know what effect surface area of PSCs has in addition to the background aerosol.

f. We cannot quantitatively assess the role of binaries in ozone depletion, and we do not claim to be able to. After all, an observed ClOx entity does not tell us if the parent Cl_2 molecule was produced on the surface of NAT, ice or a binary aerosol particle. We do present evidence that with the activation rates known from laboratory studies, the models used here (CLaMS, ATLAS) do not need the formation of PSCs to activate chlorine, and that chlorine activation can occur under conditions, where PSC particles cannot form. Again, this is notwithstanding the role of PSCs for denitrification, which we acknowledge and which is implemented in the models.

g. I think there is some misunderstanding on the conclusions that we do draw on binaries:

- laboratory based activation rates increase with decreasing temperature, and this increase is more important than the increase in surface area provided by PSCs.
- under the conditions we investigated (e.g. March 2005, winter 2010), laboratory based activation rates are sufficiently fast to drive chlorine activation (but not necessarily ozone loss as there are other factors and processes factoring in) in our models.
- there is some evidence that some of the observed chlorine activation does occur on binaries, in particular in early winter and under denitrified conditions.

Again, we do not claim that the reactions on binaries drive ozone loss or will significantly change simulated polar ozone loss for any past Arctic or Antarctic winter. The side of deactivation, and

how this is influenced by PSC formation and subsequent denitrification, is also described in the paper. In the revised version, we will highlight the importance of a correct representation of denitrification in the global modelling section, and we will start the section on heterogeneous chlorine activation with a clear and unambiguous statement that the investigations presented concern the importance for the chlorine activation process, and that the impact on ozone loss is minor (as has been shown in the Wohltman et al. paper).

28) The Kreycy et al. paper has positively been reviewed (see here http://www.atmos-chem-phys-discuss.net/12/27821/2012/acpd-12-27821-2012-discussion.html), and now it awaits a final decision from the editor. A more comprehensive study (going beyond the results reported in the paper) has been published in the PhD thesis of S. Kreycy, University of Heidelberg, 2011, which will soon appear on the web site of the University of Heidelberg/Germany (it can already be obtained upon request by email to: skreycy@iup.uni-heidelberg.de).

The novelty of the study comes with:

(a) for the first time, stratospheric O_3, NO_2, and BrO were simultaneously measured in limb (i.e. time-dependent) and solar occultation (SO) geometry using two optical balloon-borne spectrometers (beside the measurement of other more transport related tracers using a mid IR spectromter). The measurements were performed in the high latitudes stratosphere during dusk and dawn, when arguable the kinetics of the reaction BrO+NO_2 and photolysis of $BrONO_2$ are most relevant for the stratospheric bromine photochemistry, i.e. during summer to winter circulation turnover. Balloon-borne limb and SO measurements generally are (i) more specific to, and (ii) provide much larger sensitivity for detection of the targeted gases than ground- or satellite-based spectroscopic measurements (e..g., due to the better defined light paths and the larger air mass factors for the probed air masses).

(b) modelled and measured limb radiances were inter-compared in order to find confidence (i) in the overall approach and (ii) in the UV/Vis actinic flux (which were previously tested by e.g. Bösch et al., Geophys. Res. Lett., 28, 1179–1182,2001, and Deutschmann et al., J. Quant. Spectrosc. Ra., 112, 1119–1137, 2011) later used in the photochemical modelling. Here that total solar irradiance was also measured and inter-compared to satellite measurements in a one of our previous studies (e.g. Gurlit et al., Atmos. Chem. Phys., 5, 1879–1890, 2005)

(c) measured and modelled path integrals (or slant column amounts) of the relevant gases were the inter-compared using a combination of state of the art RT (McArtim, e.g., Deutschmann et al. 2011) and photochemical model (which in essence was the photochemical code of the CTM SLIMCAT). While excellent agreement is found for measured and modelled path integrals of O_3 and NO_2, measured BrO was larger (69%) than modelled using the JPL-2011 reaction rate coefficient for BrO+NO_2 and the absorption cross section for $BrONO_2$ (both for the series of limb and the SO measurements).

Further the investigations included a larger set of sensitivity studies (c.f., by varying the ratio of $J(BrONO_2)/k(NO_2+BrO)$ and the total amount of Br_y, which was however independently determined from the investigated ratio) to show for which set of parameters best agreement between the measurement and the model can be obtained for $J(BrONO2)/k(NO_2+BrO)$. It was found that for both types of measurement (limb and SO), (excellent) agreement between the measured and the modelled BrO can only be obtained when in the photochemical $J(BrONO_2)/k(NO_2+BrO)$ is assumed to be a factor of 1.69 ± 0.04 larger than recommended by JPL for 220 (+/-5) K.

29) We cannot repeat all the information from the Laube et. al study (which appeared in ACP on 8 March 2013: http://www.atmos-chem-phys.net/13/2779/2013/acp-13-2779-2013.html). However, we will present more details in the revised version and in particular discuss differences to previous work.

30) Such a statement will be added as suggested.

31) The Kuttipurath paper has now appeared in ACP with a modified title: Antarctic ozone loss in 1979–2010: first sign of ozone recovery . A clear postive signal towards ozone recovery is now revealed in this study. This will be reflected in the revised verion of the overview paper, and more details and additional references will also be added.

32) The section on modifications to CCM made during RECONCILE and how they change the model results will be rewritten.

33) At solar zenith angles typical for polar spring, the radiation amplification factor, i.e. the change in UV radiation for a given change in column ozone, does indeed increase with decreasing ozone to start with (cf. Box 2-1 in WMO, 2010), and this is the most important factor why the radiation enhancement in the southern hemisphere is so much greater even if the amount of ozone lost is similar when, for example, comparing the 2011 Arctic winter to Antarctic winters in the 1980s. This important piece of information will be included in the revised manuscript.

34) We will move this discussion forward to where the ClO dimer cycle is first addressed in detail.

35) We will clarify that PSC processes such as denitrification have a stronger impact on polar ozone depletion than whether activation takes place on binaries or PSCs. Yet, we will explain under what circumstances binary activation could factor in measurably.

36) The statement on the representation of polar ozone depletion in global models will be changed to "The representation of polar ozone depletion in global models has also been improved by implementing a more realistic description of PSCs, denitrification, and ClO_x kinetic parameters directly derived from the RECONCILE campaign and laboratory results."

38) The uncertainty in the data shown in Figure 20 has been estimated to be in the order of 20% (e.g. Harris et al., J. Geophys. Res. 107, 8264, 10.1029/2001JD000482, 2002, and WMO 2007, Chapter 3, section 3.3.3, last sentence). We will add this information to the figure caption.

Figure 21 will be replaced by a more elaborate discussion in the text, with reference to Figure 1 in Isaksen et al. (Geophys. Res. Lett, 39, L24810, 2012), which contains the comparison with satellite observations and a measure of uncertainties in the sense that it shows the inter-annual variability range of the model and observations.

C Specific replies to Jean-Pierre Pommereau

The comments by Jean-Pierre Pommereau are addressed in the general response above.
We will add a short paragraph in the introduction that defines "the exact content of RECONCILE".

D Specific replies to Rob MacKenzie

First of all, if suggesting authored or co-authored papers for citation represented a conflict of interest, many reviews would not qualify as independent reviews. While in the past I have occasionally wondered about the relevance of references suggested in reviews, the papers pointed out by Rob MacKenzie seem appropriate to cite here.

With respect to Section 4.2.2. on refractory aerosols, we fully agree with Rob MacKenzie in the sense that a clear differentiation between measurements inside and outside the vortex was missing, which indeed caused misunderstandings. The section 4.2.2 is rephrased at essential points and the repeated use of "many" is avoided. It is replaced by more direct and quantitative statements as required.

The results on bromine chemistry (currently 4.3.3) will be mentioned in the abstract and conclusions.

The papers cited in Section 4.3.4 present results that are based on measurements made during the RECONCILE aircraft campaign. We will highlight the significance in the revised version (including brief

statements in abstact and conclusions) and give more detail on differences to previous work (cf. response to Susan Solomon's comment #29).

Sections 4.4.1 and 4.4.2 stand out from the rest of the results described in Section 4 of our paper. They will be dropped as standalone Sections and more tied into the rest of the paper (cf. some of the comments in Susan Solomon's review).

The CCM Section will be completely rewritten, and the results of the evaluation will be described in more quantitative terms.

With respect to the minor comments, we will make changes according to the criticism/suggestions raised. Figure 1 will be dropped from the revised manuscript. The model name LMDz-Reprobus will be used consistently throughout the manuscript. The end of Section 4.1.1 will be rewritten: "A comprehensive analysis of SSWs in past Arctic winters also suggests profound tropospheric forcing for the observed major SSWs (Kuttippurath and Nikulin, 2012). The authors also present a statistical analysis for the past 17 Arctic winters showing that ozone loss is closely correlated to the intensity and timing of major warmings in each winter, early warmings in December-January leading to minimal ozone loss." In the caption of Figure 15, "See Wohltmann et al. (2012) for details" will be replaced by "See main text for details".

E Specific Replies to Ross Salawitch

Numbering refers to the numbering in the review. Some issues are being addressed by the general response above.

1. Most of the criticism raised here will be addressed by restructuring the paper and rewriting the introduction in a shorter and more balanced manner as described in (4) of our general points applying to all reviews. In our scientific discussion of the ClO dimer cycle, we will add the Stimpfle et al., 2004, reference, and we will make a mention of the Salawitch et al., 1989, paper in the context of our NAT rock discussion. The work by Bruce Gary on mesoscale temperature fluctuations will also be cited in the discussion of the microphysical modelling.

2. a) The temperature climatology was based on ERA-Interim reanalysis data (Dee et al., Q. J. Royal Meteorol. Soc., 137, 553–597, 2011)[1]. The sorting was based on calculated Dec/Jan/Feb mean values of polar cap (50 °N to 90 °N) temperatures at 30 hPa and 50 hPa, respectively. This follows a purely meteorological analysis with no intention for correlation with ozone loss (in which case V_{PSC} would of course be the better choice).

   With respect to denitrification, Figure 13 seems to have been slightly misinterpreted. We did not mean to say that the vertical NO_y redistribution has actually occurred between 30 Jan and March. Obviously, it must have occurred when PSCs were present, i.e. between mid December and late January, when temperatures were low enough for PSC formation as is shown in Figure 7. We will reword the main text and the Figure caption to make this clearer in the revised version.

   The use of equivalent potential temperature, $\theta_e = \theta * \exp\{L*q/(c_p*T)\}$, makes only sense if the humidity q is large enough to change the potential temperature θ. As the stratosphere is usually rather dry, θ_e reduces to dry potential temperature ($\theta_e \rightarrow \theta$ when $q \rightarrow 0$).

2. b) With respect to the ozone loss estimates by data assimilation we change
   "The results indicated that the assimilation-system vertical transport is found to be too fast, which

[1] ERA-Interim is the latest ECMWF global atmospheric reanalysis of the period 1989 to the present. Information on the resolution, the data assimilation system, the observations and the boundary forcing of the ECMWF-Interim reanalysis project can be found at http://www.ecmwf.int/research/era/do/get/era-interim.

affects the ozone reference field used for the loss estimation, and thus points to an underestimation in the ozone loss estimate….. Improving the ozone reference field, by advecting the Met Office ozone field by the ECMWF 4D-Var winds instead of the Met Office 3D-Var assimilated winds, produced results in better agreement with those produced by the CTM. These results are in agreement with other studies that indicate that stratospheric transport is improved when meteorological analyses are produced using 4D-Var instead of 3D-Var."

to

"The results indicated that the assimilation-system vertical transport is found to be too fast, which affects the ozone reference field used for the loss estimation, and thus suggests that the ozone loss estimate is too low….. Improving the ozone reference field, by advecting the Met Office ozone field by the ECMWF 4D-Var winds instead of the Met Office 3D-Var assimilated winds, produced results in better agreement with those produced by the CTM. These results are in agreement with other studies that indicate that stratospheric transport is improved when meteorological analyses are produced using 4D-Var instead of 3D-Var. We conclude that ozone loss estimates based on 3D-Var data assimilation, reported in Jackson and Orsolini (2008), are thus likely to be too low."

For ATLAS, there is indeed an obvious discrepancy between ozone loss estimates from the model and from MLS between 500 K and 650 K, and this has been an issue in the review process of the Wohltmann et al. paper that this paragraph is based upon. Briefly, both ozone loss estimates use the same passive profile. The life time of ozone between 500 K and 650 K at the end of March is on the order of months, so the discrepancy can be caused by chemistry early in the winter or by transport effects. Correspondingly, there are two plausible explanations for the discrepancy in ozone:

a) There is a corresponding discrepancy between modelled and measured N_2O, i.e. N_2O is overestimated in the model. The discrepancies in ozone and N_2O appear when the major warming sets in at the end of January. They could be explained if there was more mixing over the vortex edge in the model (i.e. in ERA Interim) compared to reality during the major warming. The gradients of N_2O and ozone over the vortex edge in this altitude region would be compatible with that. Differences in subsidence can be excluded, since more ozone in the model would imply more subsidence in the model than in reality. However, that would mean less N_2O in the model, and not more N_2O, as actually modelled.

b) Additionally, part of the discrepancy in ozone can be caused by chemistry: There is an overestimation of HCl and an underestimation of active chlorine in the model in January just in the altitude range where the discrepancy in ozone appears. That does not explain the discrepancy in N_2O, however.

A maximum of 19.9 ppt Br_y (the exact value depends on altitude and latitude) and dimer cross sections from Burkholder et. al. (J. Phys. Chem. 94, 687-695, 1990) are used in the ATLAS simulations. These parameters have not been varied in sensitivity runs for the 2010 Arctic winter. The model contains a particle based denitrification module that includes NAT rocks. The degree of denitrification is varied in two of the sensitivity runs, but the effects of the variations are small for the 2010 winter. All these and more details are also found in Wohltmann et al.,2013.

2. c) In the context of chlorine activation on cold binary aerosols, we will i) ensure proper reference to earlier studies, and ii) highlight the importance of PSCs for denitrification and thus ozone loss (cf. our reply to Susan Solomon's comment 26).

3. The section on CCM simulations will be completely rewritten based on results obtained after the submission of the manuscript. In particular, the revised version will present the ozone trends calculated for the past and future 50 years using, for the first time in a CCM, a detailed non-equilibrium scheme for the nucleation and growth of PSC particles. The new PSC scheme implemented in the CCM is directly derived from the progress realized after the RECONCILE campaign and lab experiments. For that reason, the denitrification processes modelled in our CCM runs are more properly simulated than in the

CCMVal2 simulations. Although the impact is relatively small, we show that Arctic ozone can be reproduced satisfactorily with this new model when compared to the past ozone record. Because this result is obtained with a more realistic physics than in previous CCM studies, we believe that it also increases our level of confidence in the predictions of future ozone trends by the same model.

4. While quantification of the contribution of GHG forcing to current and future trends in the atmospheric conditions that influence polar ozone loss is of course a very important issue in the context of ozone climate interactions, it is beyond the scope of RECONCILE and was not part of the project. Nevertheless, this was one of the reasons why we wanted to include an outlook at the end of our conclusions, and we will explicitly mention greenhouse gas forcing in the revised version. CCM with improved process parameters will be helpful tools to address this question in the future.

a), b), h), j), l) adjustments will be made as suggested

c) We will reword to make clear that we are referring to the warming that started end of January. The campaign continued well into the warming in an attempt to measure ozone loss rates in the vortex remnants.

d) we will include the necessary information in a table, cf. general response (5) above.

e) This is, of course correct, and a number of useful studies using empirical or semi-empirical approaches exist. We will acknowledge this in the revised version with some representative citations. Nevertheless, the objective of RECONCILE was to pave another section of the long and difficult-to-construct road towards a "physical model" (e.g. a CCM). We will clarify this and also acknowledge, that this road is still far from being completed (to acknowledge this "work not completed" was one motivation for the outlook that we included as part of our conclusions).

f) The breakup of the vortex occurred in early February.

g) We will remove "the surprising result".

i) Without doubt, the Match technique is a useful approach to estimate ozone loss (which is why two Match campaigns were carried out in the framework of RECONCILE), and the large statistics will average out uncertainties related to individual trajectory calculations to some extent. However, the results obtained during the Match flight quantify the uncertainties of such calculations, and they are probably large enough to increase the uncertainty even on the statistical ozone loss estimates in an inhomogeneous vortex (cf. Grooß et al., Atmos.Chem. Phys., 8, 565.578, 2008): the larger these uncertainties are, the more matches you will need to average them out. And while in a homogeneous vortex, a Match radius of a few hundred km will not introduce a significant error in an individual Match, this may be very different in an inhomogeneous vortex. While the statistical method will still work in a winter where this is the case, the larger uncertainty introduced by somewhat increased "mismatching" of trajectories may forfeit some of the advantages of the precise and well resolved ozone measurements over ozone loss estimates using other methods that are based on satellite observations. The latter do have higher ozone measurement uncertainty and limited vertical and horizontal resolution but therefore tracers to characterize air masses are observed simultaneously.

Because "impair ozone loss estimates by the match technique" may sound more negative than this is meant and leaves room for interpretation, the concluding sentence of Section 4.1.2 will be reworded and the description of the effect of trajectory uncertainty on Match results will be described in more detail.

k) The CLaMS simulations indicate better agreement with the more physical parameterisation. Simulations for other winters hove not been performed yet. To clarify this issue, we will add here the reference to the study by Hoyle et al. (ACPD, submitted) in which it is clearly shown that unlike the new parameterisation, a constant nucleation rate cannot reproduce the CALIPSO observations.

m) a sentence on implications for product gas injection and on remaining questions will be added, following the lines of Ross Salawitch's comment.

n) We will make the statement more quantitative.

o) The reaction $Cl + CH_4 \rightarrow HCl$ is of course important. Our use of the term "oxidation chains of CO and CH_4" for a number important reactions including $Cl+CH_4$ was not well chosen and we will reword this.

p) There are not many ozone trends studies covering the 2000-2010 period when the first signs of ozone recovery are observed. Nevertheless, we will add more details and relevant additional references.

q) + v) We will make the DFA method and its use to assess the LMDz output more accessible in the revised manuscript.

r) Figure 1 will be dropped from the revised manuscript.

s) To avoid the political aspects of this Figure in this scientific paper we will replace it by a Figure that just shows the locations of the sites that operated during the campaigns described in this paper.

t) Figure 17 does not contain any information on the consistency with field data. The information it contains is that uncertainties in cross sections are no longer dominating the uncertainty in J.

u) As stated in the response to Susan Solomon's comment # 38 above, Figure 21 (which Ross Salawhich seems to be is referring to) will be removed from the paper. The original version of this figure shown by Isaksen et al. (Geophys. Res. Lett, 39, L24810, 2012) does contain the comparison with satellite observations.