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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evidence for ice particles in the tropical stratosphere from in-situ measurements" *by* M. de Reus et al.

Anonymous Referee #2

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The paper "Evidence for ice particles in the tropical stratosphere from in-situ measurements" by M. de Reus presents upper tropospheric and lower stratospheric aircraft observations of ice clouds and interstitial aerosols located above Hector storms systems over the Tiwi Islands, north of Darwin, N. Australia. These measurements are very important and useful to other researchers as ice that looks likely to have been injected into the stratosphere by overshooting deep convection has been sampled. Stratospheric ice size distribution data of ice from overshoots has not been reported before in the literature and will provide useful information on overshoot processes that might, for example, be used in cloud models and perhaps global models in order to further study the overshoot process. The injection of ice into the stratosphere is important as it could potentially have a global moistening effect on the stratosphere, if it happens

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frequently enough. The results here show that such ice was likely to have had a local humidifying effect on the stratosphere since the residence time of some of the ice was likely to have been long enough to have allowed it to evaporate.

The paper is well presented but often fails to discuss very thoroughly the issues raised by the data presented or misses some possible explanations. The discussion on the residence times of the ice particles observed, for example, could be fairly easily taken a step further by estimating how much of the observed ice mass in the stratosphere would be likely to evaporate and contribute to humidifying the stratosphere, and how much would fall out. If calculations of evaporation are not possible then statements on what percentage of the ice would remain in stratosphere for different amounts of time (e.g. 1 hour, 10 hours, etc) would at least give a more concrete idea of the amount of moistening that might be possible. Other particular examples are the discussion of Fig.4 (where there are also some misleading statements made), the explanation of why the ice effective radius decreases with height (the possibility of simple preferential fallout from the main convective updraught is not mentioned), the explanation of the lack of tropospheric ozone and temperature signals in the overshoot events in section 3.3 (the paper greatly overstates the correlation between low ozone, low temperature and the overshoot events and more detail is needed on how mixing might account for the results) and the discussion of the aerosol results in section 3.4. The latter section in particular needs some work as it seems to miss some important information that the data provides regarding the more frequent presence of aerosols in stratospheric ice regions compared to upper tropospheric ice. Also, there is very little discussion on Fig. 9. The discussion of Figure 10 also would benefit by getting to the bottom of whether it really does suggest shattering at higher ice water contents given that shattering is suggested to cause an overestimate of the IWC by the FSSP and not an underestimate as shown. Are there other examples in the literature where shattering causes such an underestimate? The summary is also a little lightweight - indeed the abstract does a much better job of summarising the paper so perhaps this should be used as the basis of a more fleshed out summary. More detail on these points is provided below.

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Once these issues are addressed, I recommend the publication of this paper into ACP as it details some very important measurements and highlights some interesting effects within the upper tropospheric and lower stratospheric ice clouds.

Specific comments:

p. 19315 Lines 1-2 - the statement "have a high potential for humidifying the stratosphere" might suggest humidification of the stratosphere as a whole has been shown. Also, since no estimate of how much of the mass is likely to evaporate has been made, the statement should be made less strong to something like "are likely to have had a high potential of humidifying the stratosphere locally".

p. 19328 Line 1 - which model is referred to here? Plus this sentence needs more explanation as to its meaning. Line 28 - it seems likely that when considering the outflow of deep convective storms there would be preferential lofting of the smaller ice crystals in the updraught leading to a decrease in the effective radius with height - this possibility should also be mentioned.

p. 19330 Line 6 - I disagree with the statement "At higher IWC, the two measurements seem to agree much better.", since at higher still IWC the agreement worsens again. This should be replaced with something to say that the agreement is good at an IWC of around 10E-3 g/m3 but then worsens again at higher IWCs. Line 15 - some explanation or a reference is needed for why shattering leads to an overestimation of the IWC. Line 20 - the large scatter in the Brown and Francis study at low IWC is also seen in the results here suggesting that this method is unreliable at low IWC - this is a point worth making more explicitly. Line 28 - Might the discrepancies between the measured IWC and that calculated from the size distribution also be due to errors in the sizing measurements by the FSSP or errors in the FISH/FLASH measurements rather than solely the number concentrations?

p. 19331 Line 10 - there only look to be four size distributions in the top panel of Fig. 2 rather than the five stated. Is one missing? Lines 26-27 - the supersaturation might

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also be due to uplift in a gravity wave.

p. 19332 Line 11 - the ozone and temperature decreases are far from clear in events 2,5 and 6 as well as for the mentioned event 1. This statement and the follow up that this is indicative of updraft of tropopspheric air into the stratosphere should be removed since the data simply does not look to show what is stated (at least at the level of zoom in Fig. 6.). It is still possible that the mixing of tropospheric and stratospheric air occurred but left no trace of a tropospheric signal in temperature and ozone due to the mixing between the two air masses. The total water content and the LIDAR data provide much better evidence for convective injection.

Line 12-13 - "Hence, the ice crystals in the stratosphere result from overshooting convection of the Hector system". This statement is too strong based on the ozone and temperature data alone. It should be replaced with a milder statement after the mentioning of the LIDAR data and some mention of the high IWCs (which are far too high to be explained by in-situ formation of ice). A reference to Corti et al. (2008) - referenced in the paper - could be made here. In that paper the different explanations for the ice crystals in the stratosphere are examined with the conclusion that injection by overshooting convection was the most likely explanation.

Lines 14-15 - "which show the convective system below the aircraft, reaching down to the local tropopause." - surely the convective system is reaching upwards beyond the tropopause so the word "down" confuses matters. Also, the tropopause is not marked on the figure and it cannot be seen whether the lidar backscatter reaches as high as it. What is clear is that there are remnants of the convective system visible directly below where the aircraft saw the ice crystals and high total water contents in the stratosphere, strongly suggesting that the clouds had overshot into the stratosphere. So perhaps the above can be replaced with something similar to "which show remnants of the convective system directly below the aircraft at times when the aircraft was in the stratosphere and was observing the large total water mixing ratios and high ice crystal concentrations.". If possible a line indicating the tropopause height would be useful on

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figure 6.

Line 20 - "the observed ice crystals are much too large to be formed in-situ" - it is the total water mass that is most important rather than the ice crystal sizes.

p. 19333 Lines 7-10 - I think it is necessary to state the percentage of the IWC present in the stratosphere that would take more than certain key amounts of time (e.g. 1 hour, 10 hours, etc.). This should be possible given the measurements of the ice size distributions. It would provide some useful information that might be used by other researchers to estimate how much of the ice present would be likely to evaporate (given estimates of the crystal evaporation) and moisten the stratosphere.

Lines 20-21 - "show the average interstitial aerosol concentration with the selected ice clouds". This is unclear - how is the averaging done and what are the selected ice clouds - how were they selected?

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Lines 4-5 - a reference is required for the statement "particles from primary sources are always larger in size".

Lines 5-14 - the arguments here are put down in the wrong order in my opinion, which disrupts the flow of the argument. The first statement (1) - "Therefore, it is very unlikely that these particles are transported from the boundary layer within the Hector convective system" is unsubstantiated at this stage in the paragraph. Statement (1) would be better placed towards the end of this paragraph (line 14). Similarly for the statement "Therefore we presume that the ultrafine particles have been formed in the outflow of the Hector storm system" - this is only supported in the last line of the paragraph, which also helps to count against the likelihood of formation in the boundary layer (i.e. it supports statement (1) and hence it would be better if it comes before statement (1)). I propose re-writing the paragraph as: "particles from primary sources are always larger in size (insert reference). Furthermore their formation must have been recent... ...(Cur-

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tis, 2006). A likely place for the formation of the ultrafine particles is in the outflow of the Hector storm system because this region is particularly favourable for new particle formation... ...(de Reus et al., 2001). By the same argument, it is unlikely that the particles were transported directly from the boundary layer within the Hector system."

Lines 15-22 - the statements in this paragraph go too far given the evidence presented in Fig.8, especially since it is very hard to see the small coloured squares indicating the in cloud aerosol concentrations. I disagree with the first statement "The vertical profiles of the aerosol number concentrations in and out of clouds do not differ much in the troposphere" since there seem to be many heights where there were lots of out of cloud samples of air that had higher than background aerosol concentrations and not many samples of in cloud air with such higher aerosol concentrations. Again, though, I think it is hard to conclude very firmly from Fig. 8 and so I suggest that you produce separate plots of the distribution of aerosol concentrations for samples both in and out of cloud - i.e. frequency vs. aerosol concentration. This should be done for samples below and above the stratosphere, separately. Judging from Fig. 8 this would show that in the troposphere there would be a more pronounced secondary mode at higher aerosol concentrations for out of cloud samples than for in-cloud ones. Whereas, in the stratosphere, both in and out of cloud samples look to have similar secondary modes.

Then comes the question of why this should be. The paper states that the enhanced aerosol in the cloudy stratospheric samples is "supporting the hypothesis that these air masses are transported from the troposphere by overshooting convection" (lines 18-19). However, the observations of the aerosol do not support this statement since there are other possibilities for why the ice particles are coincident with higher than background aerosol concentrations. One is that the aerosols in the stratosphere were already present from e.g. volcanic eruptions, etc. and the stratospheric ice was simply injected into this pre-existing aerosol layer. This is supported by the fact that there were points at the same heights in the stratosphere with aerosol concentrations much greater than those observed at any other heights. However, given the high ice water

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content of the stratospheric air it seems unlikely that this aerosol caused the presence of the ice through nucleation. There needs to be some discussion of these outlier points presented in the paper.

This still leaves the explanation of why the higher aerosol concentration samples below the stratosphere are generally more likely to be ice free than those in the stratosphere and some discussion on this is required. One possibility is that the higher aerosol concentrations were indicative of higher IN (Ice Nuclei) or CCN (Cloud Condensation Nuclei) concentrations in the history of those samples that somehow led to the formation of larger ice crystals that sedimented out of the cloud. The generally smaller size of the ice crystals in the stratosphere might have prevented this from happening there.

p.19335 Line 11 - "this work suggests that the ratio is very dependent on the development stage of the cloud". Is this referring to the Seifert work or that presented in the paper? This should be made clear. I think that the paper needs to say a little more about the results than this - for example, it looks to me that there are two regimes in the scatter plot - one where there is some positive correlation and one where there is no correlation similar to that mentioned from the Seifert work. These could represent regions of in-situ ice formation (positive correlation) and regions where the ice crystal concentration is likely to have been determined by microphysical factors lower down in the cloud (no correlation) such as e.g. IN concentrations, CCN and droplet concentrations, updraught speeds, Hallet Mossop splintering, etc. Some further discussion on these possibilities is required even if they cannot be proved from the data, otherwise there seems little point in the plot.

Dicussion (section 4) p. 19336 Line 1 - The "At times between...." sentence is a little clumsy and would be better combined with the last statement about the cloud being patchy. E.g. "Note, however, that the cloud structure is patchy and not completely filled with crystals as suggested by the fact that, when flying at 18.8 km, no ice was measured in between the different encounters with the ice particles".

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p.19337 Line 4 - Why should the linear relation between the two IWC values indicate the occurrence of shattering? It seems to me that the relationship is approximately linear at all IWC contents, not just for >10E-4 g/m3 and that this might be expected since high IWC contents at larger sizes seem likely to be coincident with high IWC at lower sizes. Therefore, I"m not convinced that the plot suggests the occurrence of any shattering. Are there any plots of this type available for comparison that show cases where shattering is known to have occurred and where the FSSP underestimated the IWC? It should be mentioned here that the ice crystal sizes are small (as mentioned earlier on p. 19319) since it is directly relevant to the argument of whether shattering occurred.

5. Summary and conclusions The summary is generally a bit too insubstantial and misses some of things discussed in the rest of the paper. Line 13 - the statement "and might be very important for humidifying the stratosphere" is too strong and is not supported by the evidence presented in the paper (since no estimate of how much of the mass is likely to evaporate is made). This should be changed to something like "and are likely to have humidified the stratosphere locally, although no estimate of the magnitude of the humidification, nor of the likely global significance, has been made here."

Something about the other issues discussed in the paper should also be summarised - i.e. the information on the stratospheric ice size distributions; the discrepancy between the IWC calculated from the size distributions and the parameterisation and the points about the interstitial aerosol concentrations measured (after dealing with the points raised earlier in this review).

Technical corrections: p. 19314 Line 13 - insert "of" between "comprise" and "sizes". p. 19315 Line 9 - should insert "and reradiate" after "absorb" and replace "act as" with "act like".

Line 22 and throughout - use of "moreover". This word suggests that the additional

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point being introduced is more important than the previous points (like "More importantly... "). Therefore I don"t think that it should be used as a straight substitution for e.g. "In addition, ...".

p. 19316 Line 11 - the part of the sentence starting with "cirrus" doesn"t flow well and should be changed to e.g. "cirrus measurements were performed in the mid-latitudes of the northern and southern hemispheres at up to 12 km in altitude." Line 15 - the abbreviations NH and SH should be defined when first introduced. Line 28 - insert "of" after "water content" and "ice crystal sizes". Line 29 - insert "of" after "forcing". Use of "moreover".

p. 19317 Line 5 - "as an only 200-300 m thick cloud layer" sounds wrong. Would be better as "a cloud layer only 200-200 m thick". Line 21 - "Moreover".

p. 19320 Line 13 - shouldn"t be a comma after "elements". Lines 18-21 - this sentence could be a little confusing. I suggest putting a full stop after "the minimum and the maximum dimension" and then saying "These are defined as ...".

p. 19321 Line 3 - replace "over" with "to". Line 9 - the sentence starting with "This lost slice" does not explain the procedure very well. Please make it clearer.

p. 19322 Line 5 - "when it shadows two diodes completely and two for only 49.8 Line 20 - commas are not needed after "distribution" and "CIP" Line 25 - "while" does not work in this context and should be changed to "because".

p. 19323 Line 2 - add an "s" to "result". Replace "as have been observed" with "than were observed". Line 5 - replace "which" with "but".

p. 19324 Line 8 - "and only non-volatile particles to be counted". This sentence can be confusing and would be better as "and meaning that only non-volatile particles are counted". Line 23 - insert "such" before "as".

p. 19325 Line 24 - should be no commas after "clouds" and "made".

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p. 19326 Line 16 - replace "peaks" with "peak".

p. 19327 Line 11-12 - remove "which have been". Lines 16-17 - comma required after "largest mode" and "tropopause region". Lines 18 - comma required after "We have to note". Line 22 - add "s" after "distribution".

p. 19328 Line 5 - there should either be a ";" after "-40 degC" or a new sentence should be started. Line 21 - "and ambient temperature" - should be "and decreasing ambient temperature". Line 24 - similarly - should be "decreasing ambient temperature". Line 28 - replace "aging" with "ageing".

p. 19329 Line 16 - insert "decreasing" after "content with". Line 20 - "lower as" should be replaced with "lower than". Line 24 - add "s" after "hygrometer".

P. 19330 Line 7 - need "of" in between "factor" and "four". Line 14 - need "the" in between "in" and "case".

P. 19331 Line 26 - add an "s" after "period".

p. 19332 Line 1 - insert "which is" before "an area" to make it clear that what follows describes the effective radius. Line 6 - insert "of" between "factor" and "two". Line 12 - replace "for" with "of". Line 26 - replace the two instances of "its" with "their" since the sentence is talking about the ice crystals as plural.

p. 19333 Line 17 - change "concentration" to "concentrations". Line 18 - change "is" to "are".

p.19334 Line 4 - no need for comma after "atmosphere". Line 6 - change "system" to "systems" as are talking about 5 different flights. Line 15 - change "profile" to "profiles" and "concentration" to "concentrations". Line 17 - the ultrafine particles also look to show enhancement. Line 17 - commas required after "total" and "concentration". Line 17 - change "concentration" to "concentrations" and "is" to "are". Line 24 - change "crystals" to "crystal". Line 26 - change "As the three lines show the ratio between... ...ranges from..." to "The three lines show that the ratio between... ...ranges from..."

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p. 19335 Line 4 - change "concentrations" to "concentration". Line 24 - change "extend" to "extent".

p. 19336 Line 9 - insert "such" before "as". Line 10 - change "its" to "the". Line 12 - insert "with sizes" after "particles". Lines 14-15 - remove the second "number concentration" and add an "s" to "particle". Commas required around "for example". Line 17 - insert "the" after "that". Line 20 - add an "s" after "time" and a comma after "available". Line 24 - add an "s" after "value".

p. 19345 Caption for table 2 should indicate that the data is only for 30th Nov.

p. 19350 Fig 5 - this figure should use proper maps instead of the idealised coastline shown.

p. 19353 Fig 8 - the legend should indicate that the large squares are for the average interstitial aerosol to distinguish them from the smaller coloured squares. The description for Fig. 8 should indicate that "in cloud" is the same as interstitial as otherwise confusion could arise.

p. 19354 Fig 9 description - should indicate that Naerosol is the interstitial aerosol concentration and Ncloud is the ice concentration.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 19313, 2008.