The authors appreciate the detailed and thoughtful comments on their manuscripts and offer their responses below and are reflected in the returned manuscript.

Anonymous Referee #1
Received and published: 11 January 2013
1) General comments:
The manuscript by Thomason and Vernier presents a detailed new analysis of the SAGE II extinction measurements with improved discrimination between clouds or cloud-aerosol mixtures and aerosols with respect to the original so-called Kent method. This dataset is of high scientific interest, because no other long record aerosol dataset covers the 1980s, 90s and up to 2005. The paper investigates the recent finding of the Asian Tropopause Aerosol Layer (ATAL) by the CALIPSO instrument and the SAGE II results show no significant ATAL signal in the dataset before 1999. The most critical point in the analysis is the differentiation between aerosol and clouds. This procedure is described in detail but the robustness of the method is not completely convincing. The reader may think that clouds can create similar signals. Some improvements in the description of the method are necessary for final publication. ACP is exactly the correct place for the publication of the results. The paper is well written but some technical changes will improve the quality of the manuscript (see comments below).

2) Major comments:
Centroid Method: *The description of the aerosol, "artificial" cloud centroid, and the corresponding "mixing centroid function" R is difficult to understand (Sec. 2.3 and 3.2) and not completely convincing why these methods help to detect the ATAL signal:*

We have spent some time working through the description of this technique to strength its understandability. We hope that this reviewer will find it clearer in this version. The new cloud method is a part of the process to ensure that the influence of clouds on the aerosol analysis is minimized. This purpose has broader applications than the ATAL analysis but it is particularly helpful when we look at individual years prior to 2000 (Figure 10) and in multiple year sets as shown in Figures 8 and 9.

(a) *Are R and a in Sec. 2.3 sensitive to altitude and season? This should be mentioned.*

R and a are dependent on altitude and/or season in as much as the parameters of the equations are variable (R_a and k_a only, since R_c and k_c are fixed). The pdfs and the resultant values for R_a and k_a are somewhat dependent on altitude but generally insensitive to latitude and time. The heavily volcanic periods prior to 1997 are very different and cloud identification in the densest periods is difficult to impossible. We have avoided these periods in our analysis. We have added a figure that shows the dependence of dividing line between primary aerosol and enhanced aerosol/cloud and the extinction ratio centroid as a function of altitude. Text regarding this and the temporal dependence has also been added (also in response to comment below).

(*b*) *Is it possible to prove the 'mixture' line with model calculation of realistic particle size distributions and mixtures. This would give more confidence to the approach.*

I am not sure adding modeling would add to this discussion. Any aerosol model would be judged realistic if it reproduced the extinction and extinction ratio we measured with the instrument. The extinction measurements have an extensive validation history and we are confident that the measured extinction levels and 525 to 1020-nm extinction ratios are reliable (the latter roughly corresponding to an effective radius between 0.15 and 0.2 microns in figure 4/6). The notional 'cloud' is admittedly artificial (and referred to as such) but for extinction observations, virtually any particle or collection of particles with a radius greater than 0.5 microns would yield a 525 to 1020-nm extinction ratio close to 1. As a result, using a cloud extinction ratio of 1 is easily justifiable. The extinction value is more problematic but the end results are not sensitive to the notional cloud extinction coefficient as long as we use a value greater than 10-3/km.* At any point the 'cloud' extinction values cannot be taken as representative of the actual cloud extinction since we can't know how the cloud is distribution along the observational line of sight and interpreting cloud extinction coefficient is very difficult and basically represent a lower bound on the possible extinction of the cloud. An empirical mixing of cloud and aerosol is justified since the mixing is driven by geometry along the line of sight rather than mixing of cloud and aerosol within the same air. We hope that the modifications of this discussion have improved the clarity for the reviewer.

*The location of the red line in Figure 6 that divides the aerosol is almost identical (and indistinguishable on this plot) whether the cloud extinction is set to 10-3, 10-2, or 10-1/km or even greater though the fraction of cloud vs. aerosol (parameter *a*) is different for the same extinction values. Using a smaller value would generally push the curve below the one shown in the figure. Realistically clouds we observe exhibit a broad range of extinctions and using a large extinction is the most conservative approach to ensure removing as many clouds-influenced observations as possible.

(c) *I would suggest highlighting the centroid parameter Rc, kc, Ra, and Rc in Figure 4. This would also confirm why you use two very similar PDF figures instead of one, although all information of Fig. 4 is found in Fig. 6 as well.*

We have added the requested additional information to Figure 4. We considered condensing Figure 4 and 6 into a single figure but we thought that it was too busy and would prefer to leave them separated.

(d) The difference between the two centroid methods is only marginal. Please highlight more explicitly where in the PDF diagram you win the information of aerosol signals producing the ATAL signal. Why is the Kent method less sensitive to detect the ATAL signal? Please specify how many additional aerosol measurements you get from your analysis and specify more detailed the altitude dependence on R (e.g. by a figure for 14 km). Is there no dependence with latitude as well?

The new method should be recognized as a revision of the Kent method. The revision was undertaken since we (the broader SAGE group) had realized for some time that it allowed some clouds to 'leak' into the aerosol analyses. In fact, we had developed not totally satisfactory, quick-and-dirty fixes to account for the issue in the past and the new method is an effort to formalize a more robust cloud/aerosol discrimination with applications beyond the current subject. The big difference between the aerosol analyses based on the new method and the Kent method occurs in mid and low latitudes where the Kent method appears to classify as aerosol data points that appear to be cloud contaminated (the region bound by the red, green and blue lines in figure 6). While these account for only a percent or two of the total number of 'aerosol' measurements, these almost exclusively occur in low latitudes and produced obvious artifacts in the data analyses and made identification of the ATAL method impossible.

We find very little latitude dependence on the parameters used in the analysis. They are dependent on the volcanic burden observed in the 1990s (though not greatly so in the years we examine). They are dependent on altitude and it is an excellent suggestion to add a figure showing this dependence. Therefore, we have added a figure that shows the dependence of dividing line between primary aerosol and enhanced aerosol/cloud and the extinction ratio centroid as a function of altitude for the period of 1999-2005. Text describing this and the temporal dependence has been added.

(e) *What are the effects of broken clouds (Fig. 3, clouds not filling the tangent height layer) in the classification Fig. 6. I would expect smaller extinction for similar extinction ratios. But why you cannot observe extinctions smaller than 2 10−4 km−1 for ice clouds and where should I find in the classification diagram a pure ice cloud signal? Like the authors mentioned: "One concern with the analysis would be that clouds are still slipping by the analysis and artificially creating an aerosol feature." An additional validation analysis with coincident lidar or in situ measurements could clear these concerns out. Please comment why this isn't an option.*

Broken clouds (or at least ones that appear only along segments of measurement paths) are primarily agent for the arm-like shape of that protrudes from the aerosol cluster and stretches toward high extinction/low extinction ratio. The ability to identify the presence of clouds in the SAGE II data is dependent on producing a perturbation to the observed aerosol background. Sometimes this is easy such as an observation of extinction coefficient over 0.01/km with a ratio of 1 when the bulk of the observations have extinction coefficients near 10-4/km and a ratio near 4. Because of the mixing process created by the large measurement volume, there is a strong likelihood of mixing segments of air that are cloud free with others that have thin cloud material (optically dense material would almost invariably terminate a measurement). In this situation, SAGE II provides no distinction between a thin cloud that fills the tangent layer and an optically thicker one that fills only part of the measurement volume. As a result, it is not possible to infer anything about the opacity of the clouds given the reported extinction coefficient. The lower limit of identifying a cloud is driven by level at which the measurements become indistinguishable from the background aerosol. It is possible that points inside the 'aerosol' region have some cloud influence but at such a level that it no longer impacts the interpretation of the aerosol measurements. The question of where the observations reflect a 'pure ice cloud' is also ambiguous since we can't know the details of the mixing. However, it is clear that by the time the 525 to 1020-nm extinction ratio is approaching one (with 1020-nm extinction coefficient >10-3/km) the observations are effectively dominated by clouds but even there whether it is 'pure' cloud or not is unknown. I suspect that instantaneous cloud extinction levels cover quite a range of values though under most circumstances quite a bit more dense than 2 10-4/km.

Given the large measurement volume associated with the occultation geometry and its sparse sampling in latitude/longitude/time, we have found no data with sufficient spatial/temporal coincidence and spatial extent to allow quantitative assessments of clouds observations. Other than finding that observations are generally consistent, we have found it to be virtually impossible to do anything quantitative for clouds using airborne or ground-based systems.

***F**igure 8 presents one major result of the new classification method, the ATAL signal at 16 km in the global mean extinction ratio distribution. There is a surprisingly good correspondence between the ATAL signal in JJA with the corresponding season of the SAGE II subvisible cirrus climatology in the Wang et al. (1996) Plate 4 at 17.5 km. The ATAL signal is exactly found in the regions with the highest SVC occurrence rates (up to > 60%) for this season. This makes the differentiation even more difficult. Is it possible that the new method is biased by the underlying SVC occurrence or potential trends in SVC occurrence? Is the SVC analysis by Wang et al. biased by potential aerosols?*

I had forgotten about the Wang paper and the reviewer brings up an important point. Wang used an extremely simple cloud/aerosol discrimination scheme that would tend to push enhanced aerosol into a 'cloud' category. Nonetheless, there are many clouds in this region and they represent a significant impediment to using SAGE data in this part of the atmosphere. In fact, Figure 11 (now 11/12) was developed to reassure ourselves that clouds were not the source of this feature. For instance, if all of the ATAL enhancement was located at low 525 to 1020-nm extinction ratio values (suggesting the presence of larger particles) perhaps pushing toward the 'wedge' region of figures 4/6, we would be far less convinced ourselves that this is indeed an aerosol feature. That the aerosol enhancement occurs over the full range of 525 to 1020-nm extinction ratio strongly suggests that this is indeed aerosol and not cloud. Wang's paper also made use of a pre-version 6.0 which was found to have significant altitude registration issues in the vicinity of clouds such that clouds were effectively smeared from the altitude at which they occur to significantly higher altitudes, becoming optically thinner and thinner the further away from the true cloud top. This produced many false instances of SVC that no longer exist in the later versions. In the older versions, the observation of SVC above the tropopause was relatively common and in the newer versions, with the exception of PSCs, they are almost entirely gone. I have briefly mentioned earlier work impacted by previous version deficiencies in the subsection '*Interpretation of the observations*'.

***In** my opinion it would be helpful for the reader if the authors would present figures of the CALIPSO results in conjunction with the SAGE II results to highlight similarities in the vertical and horizontal structure.*

These figures already appear in other publications and we don't it necessary to include them here again. In addition, there could be copyright issues to reproduce them here.

3) Minor comments

p27535 and Fig.7: Do you need this figure really for your analysis. The analysis and interpretation on the lower altitude aerosol signals is not very detailed. You may skip figure and discussion or present some more details. I would expect signals over northern Canada as well, a region of large and frequent boreal fires.

Both reviewers commented on this and it is clearly not the focus of the paper. Using occultation data at such low altitudes is pretty tricky with not totally satisfactory outcomes in past work (at least in our opinions). We were quite pleased with how well the new cloud clearing seemed to bring out reasonable features at such low altitudes but the reviewers are correct that we should do more work either in this publication or in a future one. Since we do not wish to distract from the primary science outcome, we have chosen to remove this material from the paper.

P27540, l25: Please specify why "at least 2003" is an episodic event?

Earlier in the manuscript we suggest that it is likely to be a volcanic event. It does not develop like the ATAL feature in that while it appears in summer it strengthens into the fall (whereas other years show the ATAL disappearing rapidly) and acts in many ways like the volcanic events that dot the CALIPSO depictions of ATAL as well. We have added text to indicate this.

Fig. 8: Have you specified in the manuscript why you are using extinction ratio (relative to molecular) instead of extinction?

We find using extinction ratio in some circumstances more revealing than using straight up extinction coefficient though it shows basically the same things. Extinction ratio is as close to an aerosol mixing ratio as the instrument provides and avoids showing vertical gradients (with respect to altitude/pressure) when the aerosol is reasonably well mixed and generally allows features like ATAL stand out better. We have added text to indicate this.

The last sentence of the conclusion is a speculation and should be deleted. A "recent phenomenon" is not necessarily of "human origin".

You are definitely correct. We have significantly softened the 'human origin' conclusion to more on the order that it 'raises the possibility' that it is of human origin. We think it is appropriate to mention this possibility as long as it is clearly labeled as such.

4) Technical comments

p27523, l5: Vernier (2009) is missing in the reference section.

We have fixed the date on the reference and some additional problems that the first author perpetrated on the second author's publications…

l7: "performed" instead of "perform"

Done.

p27523, l15: "direct comparison" instead of "detailed comparison", because you are able to compare the results.

Done.

p27524, l15-26: this section of the introduction includes identical sentences of the abstract. It also discusses some final results of the manuscript (l21-26) which confuses the reader at this early stage of the manuscript. Please improve this section.

We have revised this section.

P27525: "is not a clear cut"

I think this is correct as is.

p27531/4: Formula (1) and (2) are nearly identical and (2) can be skipped. A detailed description of the intention why you use the _ offset would be sufficient.

Done. We have added more text to reflect that the offset compensates for uncertainties in the extinction measurements, particularly those at 525 nm. At 525 nm, measurements have substantial ozone and molecular contributions that often dominate the aerosol component leading to increased noise relative to 1020 nm where the signal is predominately aerosol at almost at aerosol levels.

P27537, l23: "saturation ternary solution (STS)" must be "supercooled ternary ..."

Done.

Fig. 4: delete "(when right of the green line)", the green line is only present in Fig. 6.

Done.

Fig. 6: please explain the regions IIa, IIb, IIIa, and IIIb in the caption or better in the corresponding manuscript section.

This was informational to ourselves but, since it isn't referenced in the paper (and not relevant to it), I have removed these designations.

Fig. 7 and 8: I expect the ATAL signal would be better to spot in a cylindrical projection.

We don't see any appreciable difference in the way the figures look by changing the project and will retain them in the form they are currently shown. We were not happy with the way the jpegs came out in the discussion version and we will be diligent in the ACP version that the figures are crisp without the jpeg compression issues evident in a few figures.

Fig. 11 is too small in the present form.

We have split this figure into 2 figures (1-d pdfs and the scatter diagrams) and omitted 2005 where data from August is missing. We will be careful during the type setting that they remain fully readable. We do think it is important to include the individual frames since they reflect the consistency of the change in individual years from the 1990s to the 2000s.