We would like to thank the reviewers and editor for their comments and corrections.

There were three (partially overlapping) main comments and taking these into account led to significant changes, including an expansion of the original manuscript. We discuss these three comments first, after which we answer the specific comments of the reviewers point by point. Our reply is in blue, reviewer comments in black.

1. Context and relevance

From the referee report nr 1, and in part nr 2. it is clear that not enough context was included, highlighting the scientific content of the presented result. Putting the results in a broader context was also encouraged by the editor.

Detection of aerosol type is one of the key challenges in aerosol remote sensing, as such data is essential for better modeling the direct and indirect radiative forcings and therefore of better constraining the aerosol contribution to climate change. As of now, there have been very few attempts to detect aerosol type from space, all with limited success. The fact that our approach works well is therefore very promising, and could have implications on the choice of spectral range of future aerosol sounding missions. We have greatly expanded our introduction and conclusion to highlight this significance.

The editor also pointed out the related need of putting the presented products in the context of other products. This is now also done in the introduction, but also in the presentation of the examples, which ties to the next point.

2. Comparisons

Related to the previous point, report nr 2 and 3 rightly point out the need for comparative analysis. While all presented products have been compared in detail with other data, to guarantee their robustness, this was only discussed briefly in the original manuscript.

We originally thought that a detailed comparison would bloat the already lengthy manuscript. Also, the presented product is quite unique. There are, as far as we know, no publicly available aerosol products which differentiate aerosol composition the way it is done in the current paper. For that reason, comparison is only possible in remote areas, where we can compare with non discriminative products.

As the third reviewer pointed out however, such a comparison does not need to be long and has the advantage that the reader can appreciate better the virtues of the new product. While full validation is obviously outside the scope of this paper, we have now compared the detection all five aerosol types with one of the most popular (and available) space based aerosol product, namely the MODIS AOD product. We have added 4 new MODIS figures and related discussions to the revised manuscript:

1. A four panel figure, with four scenes of volcanic tropospheric sulfate aerosols. The comparison is only good for Kilauea aerosols, where there are little other background aerosols. The fact that only the IR data can distinguish the sulfate aerosols in the three other scenes seems to be a clear demonstration of strength of the unified approach.

2. A comparison of AOD for two days with large dust plumes over the Northern Hemisphere. The comparison is highly favorable.
3. The global MODIS AOD average of June 2010. This is used to discuss the mineral detection (ash+sand) and that of ammonium sulfate.
4. The MODIS AOD average for October 2010 over the South Atlantic and Indian Ocean. This is employed for the comparison of smoke aerosol. For obvious reasons, this is the only type of aerosol for which MODIS shows a better detection.

These four new figures and the discussions surrounding them clearly highlight the strengths and advantages of the unified approach, as well as giving the reader context. We therefore think it was a very good idea to include them.

We also hope that in response to these first two comments, the revised manuscript is now more balanced across context, methods, applications and comparisons and hopefully is now fully suitable for ACP.

3. Algorithmic details and meaning of RN

The three reviewers all have specific questions regarding the algorithm and the meaning of the quantity RN. We believe that some of the confusion was due to the way the relevant section and subsections were structured and named. In the revised manuscript we have reordered, restructured and expanded the description of the algorithm. In particular we have added a short summary section of the different steps required, this to rule out any ambiguity. The current structure of Section 3 is as follows:

3. A general approach
    - 3.1 First pillar: Discriminant analysis
    - 3.2 Second pillar: Mahalanobis distance
    - 3.3 Summary of the algorithm
    - 3.4 Practical considerations

The meaning and interpretation of RN was implied in the original manuscript, via the link with the paper of Walker et al. and optimal estimation approaches. It is a pseudo column or apparent column and related to magnitude of the spectral extinction. In the revised manuscript we discuss the interpretation in great length. At the end of the discussion, in section 3.1 we conclude “the quantity RN is broadly speaking proportional to the aerosol extinction and scaled and normalized in such a way that its value expresses the distance in standard deviations from the unpolluted observations used in the training set”. While the meaning of RN is not necessarily easy to grasp, this quantity is at the very hearth of the algorithm and cannot be avoided. We have done every effort though to make its interpretation as clear as possible in two lengthy paragraphs at the end of section 3.1.

Report 1

The paper by Clarisse et al. deals with the detection of various aerosol types or ice clouds from spaceborne high spectral infrared measurements made by IASI. A substantial part of this paper consists of a review of existing detection methods (feature detection, spectral fitting, distance approaches, singular value decomposition, principal component analysis) and discusses their advantages and drawbacks. With the aim of generalizing and better understanding the above methods, a “general approach” is then proposed and described : the discriminant analysis method. This method is then used to briefly present six applications: ice crystals, sulfuric acid droplets, windblown sand, volcanic ash, ammonium sulfate, and smoke. Here or there, comments are made to answer difficulties met as, for example, in case of windblown sand for which the construction of covariance matrices is not trivial.

Concerning the applications, each of them, important in itself, is presented in a much too concise manner.

The goal of the paper is not to give a detailed analysis of case studies, but rather to give proof of concept of the method and to demonstrate the potential using convincing examples. Also we do not feel that these examples are presented that concisely. The discussion of sulfate aerosol for instance is alone the length of a GRL paper (3 pages text, 3 large figures). Note that we have also expanded the manuscript with 4 new comparison figures (see General Comment 2 above).

There is also some lack of clarity regarding the way the “general approach” as it is described in the first part is finally applied to each case. A selection of fewer examples, given with more extensive explanations, would be more convincing. As it is, the paper is largely methodological. This paper is constructive but rather technical and appears better suited to AMTD. I thus recommend the paper be resubmitted to AMTD.

We hope that section 3 is now much clearer as it has been reordered and expanded with a summary of the algorithm (see General Comment 3 above). The link with the examples should also be clearer now. Following General Comment 1 and 2 above, the manuscript has been significantly expanded both in text and figures. Doing so there is now a better balance between on the one hand the algorithm/methodological part of the paper and on the other hand the context, examples and comparisons and therefore should be of interest to the wide atmospheric community.

Report 2

This paper looks to introduce a new 'unified' detection method, linking spectral fitting, minimisation, principal component analysis and the sensible use of auxiliary information to develop a 'discriminant analysis' technique. This is designed to be applied to detect different classes of aerosol using infrared hyper-spectral observations, here from IASI, although the method could be adapted to other similar instruments.

More than half of the paper is spent reviewing the previous techniques before essentially combining these in the new framework. This part is in general very well written (bar a few missing definitions of some of the terms in equations) but unsurprisingly rather technical in nature and for this reason (and those given in the next paragraph) I would suggest that the paper as it stands is better suited for submission to Atmospheric Measurement Techniques.

We understand that the referee found the review section unbalanced. In the revised manuscript only 3 of the 25 pages are dedicated to the review. As we mentioned in the reply to report 1, following the detailed General Comment 1 and 2 above, the manuscript has been significantly expanded both in text and figures. Doing so, there is now a better balance between on one hand the algorithm/methodological part of the paper and on the other hand the context, examples and comparisons. These additions should make the paper more appropriate for ACP.

Rather less emphasis is given to the examples of the application of the new technique. The writing in these sections becomes less clear, and it would be difficult for a reader to repeat the analysis performed by the authors because an insufficient amount of information is provided. For example, 'tuning' is often mentioned but without any description of what this actually entails.

We agree that the description of the technique was missing clarity in places. Please see our reply to the General Comment 3 above. We have revised the manuscript accordingly, in particular:

- the main steps of the algorithm have been summarized in a separate section (3.3), so that this should be unambiguous. This should also make the section "3.4 practical considerations", where more detailed info is given, much more clear.

- As for the specific applications, the algorithm requires a lot of training data, such as for the construction of unpolluted covariance matrix. We have verified that this training data has been described as good as possible for all examples (time periods/areas). With this information, the analysis should be fully reproducible. In the case analytical Jacobeans were calculated, all relevant parameters (size distribution, refractive index) were also provided.

- The tuning which was mentioned is only needed when a binary classification is needed (e.g. sand/no sand). Detection is achieved by comparing the quantities RN and AN to predefined thresholds. As explained in the revised manuscript, these thresholds should be chosen to maximize sensitivity and minimize false detections. Tuning then consists of looking at global maps of RN or AN, both clean and polluted with aerosol, and adjusting the thresholds until these are found suitable.

There is also, in my opinion, a lack of independent evaluation of the success of the approach. While for some aerosols such as ammonia sulphate this might be difficult to obtain, for windblown sand, smoke aerosol and volcanic ash there are plenty of existing datasets that could be used.

We hope to have adequately addressed this comment now with the inclusion of MODIS AOD comparisons. Please see our reply to the General Comment 2 above.

The additional merit that this new approach brings above and beyond what can be done already is not, in my opinion, adequately demonstrated at the moment. It may be easier to do this if the authors focus on fewer aerosol types - perhaps pick those where this approach offers the biggest potential for improving our knowledge of the global distribution – and describe exactly what they have done and what the benefits are. I would certainly lose cirrus clouds as considering these as an aerosol seems rather odd to me if not technically incorrect.

Please see again the reply to the General Comment 1, 2 and 3 above. In particular we have significantly expanded our introduction, underlining the significance and potential of the presented result. It is hopefully also sufficiently clear now that the current product is unique in terms of aerosol type detection, with respect to state-of-the-art shortwave sounding techniques. The comparisons with MODIS also underline this. While the method itself should be clear now; we have as you suggested removed the discussion of ice particles/cirrus clouds, as this example didn't contribute much.

A few small additional comments:

I think it would be highly beneficial to explain to the reader what the range of RN actually means in the various figures if the authors decide to stick with this representation.

Two lengthy paragraphs have been added, explaining the interpretation and range of RN in detail (see General Comment 3 above)

In the ash and dust sections, I think it would make more sense to give an indication of where the various categories (1-10) come from geographically.

This is unfortunately not feasible as the data was clustered based on spectral signature, and not on geographical location. While nearby observations from the training data were often clustered together this is not always the case.

There is no real discussion of the effects of variations in aerosol size distribution. This, in combination with the chemical composition, will influence the optical properties that then propagate through to the radiative signature. Are the authors convinced that this will be a secondary effect? A similar comment could be made for particle shape.

For the aerosol types we have studied, these are indeed secondary effects as witnessed by the many comparisons. The importance is perhaps largest for ice particles, for which the Mie approximation is not very good. Following also your comment above, we have now removed this 'aerosol' type. The effects you mention could also be important for mineral aerosols, however variability due to particle size or shape, is partially taken into account as we work in a range of different sand/ash classes.

I wonder whether the separation used in practice into ’clean’ and ’polluted’ cases gives enough samples to exclude the impacts of variations in, in particular water vapour. While the atmospheric window is less influenced by this gas than other spectral regions, insufficiently characterising its effects will alter the precise spectral signature seen here.

The construction of the covariance matrices was typically done with millions of ‘clean’ spectra, during all periods of the year. Effects in the atmospheric window due to variations of water vapour, ozone, and other trace gases are therefore fully accounted for in this covariance matrix, and as a result in the detection.

Similarly, when calculating covariances, is any account taken of the fact that the presence of aerosol is likely to change the thermal structure of the atmosphere?

The assumption that the ‘clean’ and ‘polluted’ covariance matrices are the same, is an approximation. However, this directly follows from the assumption that the probability density function of moderately polluted and clear spectra differ only in their mean and not in their covariance. As we have argued in the manuscript this assumption is very reasonable. While aerosols will have some impact on the thermal structure, this will be a secondary effect on the spectral signature. Also if the Jacobeans are calculated from observed spectra, such effects are taken into account.

Report 3

Overall, this is a very well written and interesting paper. It provides background information about approaches that have been used for aerosol retrievals, and discusses how the different approaches relate to one another. After providing background information, aerosol analysis is applied to IASI data, and results are reported for sulfuric acid aerosols, volcanic ash, sand, ash, smoke, and ammonium sulfate.

We would like to thank the third referee for the detailed and constructive review. The comments, addressed below, have certainly helped in improving the manuscript.

major comment 1: The paper begins with a section that talks about techniques that have been applied to the problem of detecting clouds and aerosols in remote sensing data, with an emphasis on the infrared. This is a very good survey of the literature and a well written section. The authors have taken care to show the connections between the techniques and how the equations relate to one another. I think the paper needs an additional paragraph or two, that ties the wide range of methods described to the actual techniques that are employed in the paper. One can try to infer it, based on the data that is needed (covariance matrices, etc), but it would be a much clearer paper if the authors simply add a paragraph that says, we apply the following technique for aerosol analysis. In addition, there are two topics that need to be addressed in relation to the interpretation. There are when are the values of Rn meaningful, and how to apply error analysis.

We have addressed this comment fully in the revised manuscript, please see the General Comment 3 above. Note that we now also discuss the meaning of the quantity Rn, when the values are meaningful and the expected uncertainty.

major comment 2: The authors present most of the results in terms of R sub N, a distance quantity. Due to the noise on the measurement spectra, it seems there is a distance quantity that is meaningful, and some level below which the aerosols can not be differentiated from noise. This issue of when species are detectable and when they are masked in the noise is not directly addressed, and the maps and timeseries present data over a wide range of R sub N values. The maps tend not to have color bars below 0.5, but the sulfuric acid aerosol timeseries (figure 3) shows values that appear to be negative. If I understand equation 14 correctly, these are normalized values, and for clean spectra there is a mean of 0 and a std dev of 1. But most of the maps typically R sub N values of 0.5 to 2.5 or 3. Please add a few sentences to help the reader understand the expected values and how to interpret them.

major comment 3: Many of us are used to remote sensed quantities that are in physical units - optical depths, DU, or total columns. I realize that R_n is a different type of quantity, but it would be helpful to know how it relates to these other quantities – can the authors provide some indication of sensitivity - how does R_n change with optical depth for a particular species of interest. I would hope that this has been assessed some simulations analysis. This is also important because other data sources are used in comparison, so it is helpful to know how R_n relates to AOD or DU or other commonly reported quantities.

These two comments are now fully addressed (see General Comment 3 above). In particular we explain that RN expresses the distance in standard deviations from the unpolluted observations used in the training set. The expected value on unpolluted observations is 0 +/- 1 (hence can also be negative). A value of 3 almost always means confirmed detection (and 99.7% of unpolluted spectra will have a value of RN within +/- 3), definitely when such a value is obtained from averaging. We also explain the interpretation of RN as an apparent column, and under what conditions it is linearly related to the true column.

major comment 4: In a paper that reports the results of techniques for identifying aerosols in infrared spectra, it is critically important that comparison data is also presented. In this paper, the authors present detections of ice crystals, sulfuric acid droplets, windblown sand, volcanic ash, ammonium sulfate, and smoke. In some cases, other data sources are cited. for example, the discussion of sulfuric acid aerosols, papers about OSIRIS measurements and ACE measurements are cited, and claimed to be consistent. For ammonium sulfate, some AOD (not species specific) is referred to as correlative data. The sand case does not talk about any other measurements data - was MODIS and MISR data looked at - if so, was there evidence of the same dust storms, or no evidence of dust? The authors claim there is no global dust product to compare to, but certainly MISR reported large, non-spherical particles are relevant to this problem. A quick look at MISR maps suggests that they see similar patterns (see http://eosweb.larc.nasa.gov/PRODOCS/misr/level3/level3_CGAS_small.html)

We thank the referee for the suggestions. We have now introduced (MODIS and in one case also OMI) AOD comparisons throughout the paper, see General Comment 2 above. We chose this product because of its widespread use and public availability. In particular, we have added 2 comparison figures and related discussions for dust.

The ice crystal case is also problematic. It may be simply an issue of graphics - when I look at Figure 1, the large swaths of IASI data appear as red and mostly gray blocks. Some of the regions that are more gray, indicating a higher cloud fraction in EUMETSAT data, also have some hints of color that indicate a range of Rn values above 1. As I asked earlier, if the Rn value is low, are these meaningful detections? Perhaps this figure would be easier to read if only data above a certain value is shown. Alternatively, perhaps only show data if Rn is above a certain value when the EUMETSAT cloud fraction is above a certain threshold?

As requested by the second reviewer we have now removed the ice crystal case from the paper.

The volcanic ash data are also shown alone, with no other correlative data. Is there any information from the volcanic ash advisory that could be used to compare and contrast with the IASI results?

The volcanic ash algorithm has since the submission of the manuscript been run on over 5 years of IASI data. The result are very encouraging, both in terms of the number of real and false detections. Where possible we have compared this data to published material, and found an overall excellent match. We have added a few lines to indicate that such a comparison was carried out and that a good agreement was found.

Again, for the smoke case, there must be some MODIS or MISR data, at least close to the source, that could be used to help corroborate the IASI measurement. For all of these cases, a long paragraph is not required - just a couple of sentences to provide context and indicate that other data sources were thoroughly examined and compared to the IASI measurements.

The smoke case has now been compared and discussed with MODIS data.

comment 5: page 26884, lines 5 through 8 I'm confused by the sentence "For increasing aerosol loadings the covariance matrix will have a component due to aerosol covariance, but no problems are expected here, since these are easier to detect anyway" Are the authors saying that high aerosol loading cases are not consistent with the assumptions. But, this is not a problem because they can be easily detected? If that is the message, does it imply that a different approach will be needed for high aerosol loadings? Overall, this sentence is not clear or convincing.

Yes high aerosol loadings will probably not match the Gaussian normality assumption. However this does not affect detection at all. The detection is hardest at the boundary of the clean/polluted observation sets, where this assumption is most critical. Observations with very high aerosol loadings are very far from that edge, where the assumption is not critical, i.e. there are very easy to detect in the current scheme. This becomes also evident from looking at the volcanic ash test case where detection was never a problem in the plume centers. We agree that the sentence was not sufficiently clear. We have rephrased it as: "For increasing aerosol loadings the covariance matrix S_p will have a component due to aerosol covariance, but no problems are expected here, since the normality assumption is not critical for the detection of their large signature."

comment 6: page 26886, lines 8-14 The authors describe a technique where they perform a detection test for number of subgroups, and design the subgroups based on spectral clustering. Since this paper has a focus of describing a wide range of techniques and showing how they relate to one another, I am interested to know if this approach is related to the discrimination analysis approach described earlier.

As far as we know, there is no direct relation. K-means clustering is however related with the AN quantity we use, as they both employ the Mahalonobis distance as a distance criterion.

comment 7: page 26889 line 24 in addition to ISAMS observations of sulfuric acid - water drops, the ATMOS high-resolution solar occultation spectra were used to identify sulfuric acid aerosols and their composition (see papers by Steele et al, and Eldering et al.)

Thank you for bringing these to our attention, they have now been included.

comment 8: page 26891, lines 7 to 14 This paragraph talks about the detection of SO2 and H2SO4. Related to my earlier comment - the discussion says that the sulfuric acid aerosol continues to be detectable for many more months. When it the signal considered to be not detectable? Is there an Rn value that corresponds to fitting the noise, so levels below that are considered not detectable? I would like to see some quantitative information in that comment.

We have now added this information " … and it continues to be detectable for many more months after the eruption, with average RN values over 0.25 until December 2009". This value of 0.25 is obviously arbitrary, but for monthly hemispheric averages, it seems a good cut-off value for positive detection.

Minor comments:

page 26877, line 11 The authors write "With these caveats in mind, the discovery of the pseudo-retrieval method: : :.." I don't think 'discovery' is the right word here, it is implies a new understanding or revealing something that was previously hidden. I would suggest they replace 'discovery' with 'development'

Corrected

page 26880, line 7 - the use of matrix I is slightly confusing, as it is regularly used as the identity matrix, and I don't think that is the intention here. I would suggest that a different letter be selected, perhaps P, since this is related to the pollutant.

In fact, the I was used to refer to the identity matrix. We have now clarified this in the text.

equation 10 - the subscript d is never defined.

Corrected

page 26883, line 17 correct grammer current sentence: "We will not use any of techniques of mulitclass discrimination here" suggest " We will not use any multiclass discrimination techniques here" or "We will not use any of the techniques of multiclass discrimination here"

Corrected

page 26884, line 1 correct grammer now 'except to the specific', should read 'except the specific'

Corrected

page 26885, line 10. correct grammar - currently reads "to have a better grip on the quantities" suggested rewrite "to more easily interpret the quantities" or "to make the quantities more useful"

Corrected

page 26889, line 16 correct grammer: currently reads "were long enough in orbit" change to "were in orbit long enough"

Corrected

page 26890, line 22 correct grammer currently reads "we must be careful for potential spectral interference" I suggest "we must be careful of potential spectral interference"

Corrected

page 26891, lines 15 - 20. Have the authors verified that the other peaks in SO2 correspond to volcanic eruptions? A sentence or two explaining what the other peaks are caused by would be helpful.

Yes, these were verified in detail and all were found to be volcanic. A sentence has been added to clarify this.

page 26892, line 11 A map from OMI is included in as an inset in Figure 4. First of all, what is the relationship of the DU values reported by OMI and the Rn reported here (this relates to the earlier question of how Rn changes with increasing optical depth or loading: : :.) Secondly, the extend of the OMI signal is much smaller than the IASI map. Is this because of the different sensitivity of the UV and the IR, or because less data is included? A sentence or two describing the relationship or connections of these two datasets are needed if the OMI data is to be introduced.

The OMI DU refers to SO_2, while RN refers to H_2SO_4. These concern different species (and different phase gas/solid). However SO_2 is a precursor from H_2SO_4 and we indeed see evidence of this in the plots. Ie, small short-lived SO_2 plume which goes over in a larger and longer-lived H_2SO_4 plume. This was the main point of showing these maps. We have now added a sentence clarifying this.

page 26893, line 11 correct grammar currently reads "were selected directly over, and transported from the: : :.." I would suggest "were selected directly over, and over the region where the dust is transported from the: : :.."

Corrected

page 26893, line 24 correct grammar currently reads "as a better sand detection allows constructing a better: : :" I would suggest either "as a better sand detection allows us to construct a better: : :" or "as a better sand detection allow for constructing a better: : :."

Corrected

page 26893, line 25 correct grammar currently reads "to better accommodate for surface emissivity effects for the detection" I would suggest "to better accommodate for surface emissivity effects in the detection" or to better accommodate surface emissivity effects in the detection"

Corrected

page 26894, line 11 change ’in case of detection’ to ’in cases of detection’

Corrected

page 26897 line 3 the grammar needs to be corrected - currently reads "The corresponding detection thresholds were chosen quite relaxed" I suggest : The corresponding detection thresholds were chosen to be quite relaxed"

Corrected

Figure 5: The grayed out bands (which I assume are the ozone absorption band) are not explained in the text or the figure caption.

Yes, it indeed refers to the ozone absorption band, we have added this information in the caption.

Figure 7: The label of the color bar of the upper panel is missing a ')' - currently only has: Sand (%

The original figure in fact had the ')' but this had gone lost in the conversion to acpd format. It is present in the revised manuscript.

Title: I would suggest "A unified approach to infrared aerosol remote sensing and type specification" because the fact that this work is using infrared wavelengths makes it unique, so that should be brought out earlier in the title.

Corrected