We thank the two anonymous reviewers and T. Nishizawa for their careful reading and thoughtful comments, which we hope have led to a stronger final manuscript.

Anonymous Referee #1

This paper describes a method for separating external mixtures of two aerosol types using the intensive parameters (lidar ratio, color ratio, depolarization ratio) obtained from high-spectral-resolution lidar (HSRL). The method was applied to the NASA airborne HSRL data. The subject is suitable for AMTD, and the paper is well written.
The followings are questions and comments.

Is it straightforward to extend this approach to handle external mixtures of three aerosol types?
From the standpoint of the theoretical equations, yes, absolutely, the theory can handle external mixtures of three or an arbitrary number of types, and the theoretical development in the final manuscript has been extended to reflect this. The application of the theory to the separation of three types in real data is possible if the types occupy sufficiently distinct regions in the space defined by the measured intensive variables. For example, a case with dust (large depolarization), smoke (small depolarization, large lidar ratio and backscatter color ratio) and marine (small depolarization, small lidar ratio and backscatter color ratio) would be a good candidate case study. On the other hand, other combinations with less contrast (for example, smoke plus urban plus marine) would probably be more difficult. We did not include a three-component case in the data section of the paper because we felt these two-component mixtures were very clear and easy to visualize and therefore provided good support for the theory. We will work on the separation on a three-component system as part of ongoing studies.

How the pure aerosol types were defined in general? It should be mentioned if studies like cluster analysis have been conducted with HSRL-1 data.
Standard HSRL-1 data products include the results of aerosol classification following the methodology introduced by Burton et al. [2012]. That paper describes an aerosol classification study on HSRL-1 data in significant detail. We did not wish to overburden the current manuscript with a redundant description, but it appears that we erred too much on the side of brevity. We have added a more complete description of the standard aerosol classification in section 2. The standard classification results from the 2012 methodology were used to aid in the selection of pure type samples for the current study. However, the multi-dimensional normal distributions (i.e. the pure-type "models") for this study were created for specific samples relevant to each scene rather than use general models that might include measurement samples from different conditions. To be more specific, as discussed in Section 5, the pure dust model used in the Mexico City dust+pollution example was created from a sample of local dust, rather than from the totality of all HSRL-1 measurements of dust, which also includes a significant amount of transported Saharan dust with properties that are somewhat dissimilar. The local Mexican dust and transported Saharan dust have lidar properties similar enough that the

standard HSRL-1 aerosol classification has no trouble classifying both types as dust, but different enough to affect the mixing calculation in this study, as discussed in Section 6.

Nishizawa et al., JQSRT 2010 is a better reference instead of Nishizawa et al. 2010 in Atmos. Res. They also extended their method to include independent extinction (or lidar ratio) measurement in Nishizawa, et al., IEEE Trans. Geosci. Rem. Sens, 46(12), 4094-4103, 2008 to partition optically absorptive aerosols.
Thank you. The reference has been changed.

Anonymous Referee #2

This manuscript presents a technique to determine the partition of the single aerosol types in a two-type mixture to the total aerosol backscatter/extinction coefficient by lidar remote sensing. Although the separation of the different aerosol types and their fraction to the aerosol mixture by lidar optical properties is not new itself the paper provides some new aspects to that topic. The paper is well written and interesting to read.

The authors should address some (minor) points and questions before final publication:

Page 8270, line2: Are there no better methods for aerosol source attribution?

The reviewer is referring to the first sentence of the abstract that says "Knowledge of aerosol type is important for source attribution and for determining the magnitude and assessing the consequences of aerosol radiative forcing." Yes, there are other ways of attributing aerosol sources in particular cases, such as when in situ data are available, but in situ measurements are relatively sparse. Source attribution can also be attempted using modeling, but models must be validated with data. Even when in situ and models are both present, aerosol classification from correlative remote sensing measurements can still add value (as in de Foy et al., "Aerosol plume transport and transformation in high spectral resolution lidar measurements and WRF-Flexpart simulations during the MILAGRO Field Campaign", ACP 2011). So, while it is true there are other methods of aerosol source attribution, it's not unreasonable to say that aerosol typing of remote sensing data is useful and important for this kind of knowledge. In response to the reviewer's comment, we have slightly reworded the first sentence of the abstract to deemphasize this item, saying, "Knowledge of aerosol type is important for determining the magnitude and assessing the consequences of aerosol radiative forcing, and can provide useful information for source attribution studies."

Page 8271, line6: As this sentence is not restricted on HSRL-1 measurement the Reference to Gross et al., 2013 and/or Gross et al., 2011 should be added.

Good point. A reference to Groß et al. 2013 has been added to the revised paper.

Page 8271, line 11: Give a reference for the statement that aerosol layers are frequently mixtures (e.g. Ansmann et al., 2011, Tesche et al., 2011, David et al., 2013, : : :)

We have added several references in the revised manuscript.

Page 8272, line 25-29: General question for better understanding: Does the multivariate normal distribution of the (pure or mixed) aerosol type automatically link the 'newest measurement' to the right type/mixture or is there also a kind of threshold defining the 'beginning' of one mixture and the end of another?

If we are interpreting this question correctly, the reviewer is asking about the methodology for choosing the mixing ratios most appropriate to describe a given measurement point. We think we probably made this concept more confusing than necessary by dealing with discrete values at 10%-intervals for the backscatter partition, so in the final manuscript, we have instead reported partitions on the continuum between 0 and 100%. The process for estimating the mixing amount is to minimize the Mahalanobis distance as a function of backscatter (or extinction) mixing ratio. Each backscatter mixing ratio is associated with a specific vector mean and covariance matrix (that is, a multi-normal distribution) as given by Eq. (30)-(32). The Mahalanobis distance between a given multi-normal distribution and the measurement vector is a measure of how similar the measurement is to values within the distribution. Minimizing this metric gives the best estimate of the backscatter mixing ratio. It is true that a range of multi-normal distributions overlap such that a given measurement can be consistent with a range of backscatter mixing ratios. For that reason, in the revised paper, we also give an uncertainty estimate (see below).

Page 8273, line 8: Please give the references of these earlier papers.

This line is at the conclusion of Section 1. We meant the earlier papers discussed throughout section 1, which is quite a long list. In the final revision we have changed "the" to "these" and added some of the references explicitly, like this: "expanding on the work of these earlier papers (especially Leon et al, 2003; Sugimoto and Lee, 2006; Burton et al., 2012)." By highlighting these three papers in this particular sentence, we do not mean to dismiss the other papers that have shown aerosol classification work which we acknowledged elsewhere in the introduction.

Section 3: The idea of linearizing the equation for the different intensive optical properties for the mixture is a great idea; however, the main equations defining the intensive optical properties of aerosol mixtures used in this section are not particular new. The equations are based on the same heritage as Gross et al, 2011a/b (using extinction-to backscatter ratio and particle linear depolarization ratio for aerosol type separation), Gasteiger et al., 2011 (giving the basic formula for calculating the extinction-to-backscatter ratio and the particle linear

depolarization ratio of aerosol mixtures) and Tesche et al., 2009/2011 (using particle linear depolarization ratio for quantifying the fraction of the single type to the aerosol mixture). Equations can easily be calculated from those former publications to the one building the base for the linearization in this publication. A link to those former publications should be added.

We certainly agree that the main equations (before linearization) are not new. We felt it was very important to acknowledge that our equations are based on older work and differ in two ways, the linearization such that all the equations are in the same linear form, and the generalization to multi-normal distributions. In fact, the linearization is important only insofar as it allows for the generalization to multi-normal distributions, and we would not imagine it would be enough to earn publication by itself. The references to the papers by Groß et al., (2011, 2013) , Gasteiger et al (2011), and Tesche et al. (2009) were already given in the introduction. However, our work is not based on the work of Groß, Gasteiger, and Tesche et al. As we noted in the introduction, treatment of these equations in the literature first appears in earlier papers by Leon et al. 2003 (for the lidar ratio and backscatter color ratio) and Sugimoto et al. 2003 (for depolarization), and it was these papers we used to develop our own work. We feel that we were very clear in acknowledging that we followed the derivations first given in Leon et al. 2003, Sugimoto et al. 2003, and Sugimoto and Lee 2006. We also acknowledge that later researchers (the ones you mentioned) used these equations and developed them specifically for aerosol classification work, but the introduction seems like a more fitting place to discuss this contribution, and that is where we have discussed it. In contrast, in the derivation section, what seems most appropriate to us is reference to the works where these equations were first derived and on which we rely heavily for our own derivations.

Section 4 (page 8281, line 15-16, page 8282, line 19-21): As I understand the author uses different mixing ratios for the different measurement dimensions / variables. Should not the mixing ratio be the same for all used variables when regarding the same aerosol mixture?

No. We were not careful enough about terminology and this has caused some confusion. It's true that the "backscatter partition", which is the fraction of aerosol backscatter at 532 nm attributable to one of the types (as described in Eq 2), is unique for a given aerosol mixture (this is also true of the extinction partition, although the backscatter partition and the extinction partition are not equal). However, at the lines indicated by the reviewer, we were referring to the coefficient in a linear equation such as Eq. 28 and in this case the answer is definitely no. As shown in the derivation of the equations, the mixing ratio (i.e. linear coefficient) cannot be the same for the same quantity at two different wavelengths, for example. This is an important point because assuming each dimension must have the same mixing ratio (i.e. linear coefficient) could seem intuitively plausible but is incorrect. If the mixing coefficients were the same for

each dimension, the mixing curves would always be straight lines. The development of the equations in section 3 is meant to show that the mixing ratios are not in general the same for each quantity but that the relationships between the mixing ratios are known. These relationships are derived in section 3 and summarized by Eq 31. In the revised manuscript, we will be more careful to make this distinction.

For Figure 1, we intended to illustrate the mixing of covariance matrices in a very generalized manner not specific to aerosol mixing. We arbitrarily chose three very distinct linear coefficients for the three dimensions simply to emphasize in the resulting figure that the equations do not require identical mixing ratios in each dimension.

Equation 33: Variables and indices are not defined.

This has been corrected.

General comment to Section 5-7: The authors compare their values for dust with former findings, but they do not compare the findings for other 'pure types' with former findings. Perhaps some links can be added as well. Further a reference to Esselborn et al., 2009 and/or Tesche et al., 2009 should be added for fresh Saharan dust close to the source region.

We have done so in the revised paper. However, we wish to emphasize that there is significant variability within a given type (e.g. dust properties vary with source region; smoke properties change with age), so the measurements in different locations cannot be expected to exactly agree.

Page 8286, line 25: A reference for the loss of large particles during transport should be given here.

Added.

Page 8287, line 11: The link to model and aerosol optical properties is missing. Is not the measurement accuracy (in addition to the variability of the optical properties for the pure aerosol types) a main point for uncertainties in mixing ratio and partitioning results? The impact of measurement uncertainties and overlap of the multi-normal distributions for different mixing ratios on the partitioning result should be discussed in more detail.

Yes, the reviewer is correct that the overlap of multi-normal distributions necessarily means that a measurement is consistent with a range of different partition values. We have revised our calculations to also include an estimate of the uncertainty, and revised the text to discuss different error sources. The revised text is copied below:

> … A given measurement point can be consistent with a range of overlapping mixture distributions. We therefore also calculate an uncertainty in the mixing ratio estimate. The error in the extinction mixing ratio is affected by several factors. Errors in selecting or characterizing the pure type distributions would have a significant effect on the extinction mixing ratio. A large error in the pure type distributions would be noticeable as a significant mismatch between the curvature of the data and the ellipses in Figure 3; however, a smaller systematic error of this type may not be obvious and would be difficult to characterize. Measurement error in the pure type samples affects the size and shape of the ellipses but not the curves that link them, and so does not have a significant effect on the extinction mixing ratio. However, measurement uncertainty also affects the placement of the point to be characterized and will therefore lead to random error in the extinction mixing ratio. To estimate this error, we note that the minimized Mahalanobis distance is essentially a fitting residual. In order to convert the unitless Mahalanobis distance to mixing ratio units, we estimate a local scaling factor, using the Mahalanobis distance between the mixture distribution and the center of a neighboring distribution with a slightly different extinction mixing ratio. Extinction mixing ratio uncertainties for this example are approximately 3-10% mixing ratio (percentage points).

Page 8287, line 11-13: If the ellipses would not line up well with the data, is it not possible, that the mixture does not consist of the two assumed aerosol types, or that the mixture consists of more than two types?

I'm confused by this question. In the manuscript, we say that if aerosol models from the wrong pure types are used then the ellipses would not line up with the data. So, yes, we agree that if the ellipses don't line up it's because the mixture doesn't consist of the assumed types – that's exactly what we were trying to say. We were also intending to make the point that the fact that the ellipses do indeed line up with the data in Figure 3, Figure 7 and Figure 10 indicates that the pure types are well chosen and the mixture is well characterized. In addition, we were making another point that "pure types" is a category that can be interpreted too broadly. There are different kinds of dust and applying an aerosol model of transported Saharan dust to a case of locally generated Mexican dust would lead to more error in the result. In an attempt to avoid confusion, we have reworded the selected paragraph like this:

> Once again, the HSRL-1 measurements lie on a continuum between the two pure types and are in good agreement with the ellipses representing mixture covariance types from Eq. (30)-(32). This alignment indicates that the observations are well described as a mixture between these two pure types. Note however that the pure types have been specified explicitly for each scene. In particular, there is a significant difference in lidar ratio and backscatter color ratio between the pure dust samples from the Mexico scene and the Caribbean scene. This should not be surprising, since other researchers (Esselborn et al., 2009; Schuster et al., 2012; Mamouri et al., 2013) have found that the lidar ratio for dust depends on source region, and that the size distribution and angstrom exponents change as large particles are removed during transport

(Maring et al., 2003; Weinzierl et al., 2011; Preißler et al., 2013). If generic aerosol models were used in the mixture calculations, the results for the mixed state would be more approximate. The accuracy of the partitioning results depends on the accuracy of the models used. If the pure dust sample from Mexico City were used in place of the Caribbean dust model in this scene, the ellipses would not line up well with the data, which would serve as an indication that the Mexico City dust model is not a good model for this scene of transported Saharan dust. For some applications, generic aerosol models may be unavoidable, but such models would be expected to produce only approximate results for the mixed states. Further study is required to determine how to best use generic models for specific applications, and how much effect they would have on the accuracy of the results.

Page 8288, line 1-20: The papers of Sugimoto and Lee, 2006 and Tesche et al., 2009 deal with spherical and non-spherical and dust and non-dust particles, respectively. But the studies of Gasteiger et al., 2011, Gross et al., 2011 and Tesche et al., 2011 use general formulations for the mixing problem. The former two papers also use multi aerosol parameters, the latter describes the partitioning of maritime/marine and smoke aerosols and is therefore well comparable. A link to those studies should be added.

Gasteiger et al., 2011 indeed discuss equations for lidar ratio and angstrom exponent as well as depolarization ratio. They do not give equations for mixing but only say mixing is achieved "by adding the size-integrated extensive properties". The relationships are used for modeling aerosol ensembles. Instead of equations, the results are given in tabular form for aerosol models made of various ensembles. The paper is called "Modelling lidar-relevant optical properties of complex mineral dust aerosols." Because of the focus of this paper on dust mixtures, all of the tabulated ensembles are dust ensembles.

Unlike Gasteiger et al. 2011, Gross et al., 2011 give theoretical equations for lidar ratio and depolarization ratio for mixtures (but not angstrom exponent) and then discuss a means of separating out different aerosol components using these equations. But the separation technique depends greatly on the depolarization and the two mixtures that are separated in this work are both dust mixtures (marine + dust and biomass burning + dust).

Tesche et al., 2011 show a figure that features three theoretical two-component mixtures including a mixture of smoke and marine. However, the study focuses not on this type of mixture, but on a case of smoke plus dust, separating the types for the purpose of deriving the properties of pure smoke. They explicitly say that the technique depends on using depolarization and point out that the separation would be more complicated if there was little contrast between the particle depolarization ratios of the pure types (i.e. smoke and marine).

In short I would say that the equations given in these three papers are certainly general but none of them show how to apply them practically to a case of a mixture that does not include dust, which was our motivation for including the mixture discussed in section 7.

Page 8289, line 6: What is meant by 'scalar values' at this point?

We meant "single values". Equation 30 (and also the equations given by Leon et al., 2003; Sugimoto and Lee, 2006; Tesche et al. 2009, 2011; Gasteiger et al. 2011; and Gross et al. 2011) gives a single value of each lidar observable for a given pair of pure type values and a given mixing ratio. However, Equation 32 gives the distribution (not a single point) of each lidar observable for a given pair of pure type distributions and a given mixing ratio. We changed the text to say "The equations for each observable can be written in the form of a linear combination of pure types, from which follow equations for multivariate covariance matrices. Therefore we can precisely describe mixing rules not only for single measurements but also measurement distributions," which is more accurate and hopefully clearer that what was written before.

One last general question: How do the authors assign the measurement to the different mixing ratios? This is not completely clear.

I'm sorry it wasn't clear. The description of how this is done is given in Section 5. We have expanded and revised it to help with clarity. Note that we have also switched to calculating a continuous partition value instead of a discrete one as mentioned in the answer to an earlier question.

> The next step is to estimate the partitioning between the two aerosol types, in terms of the 532 nm extinction partition (or "extinction mixing ratio") for the entire flight at all altitudes. Given measured values of three aerosol intensive parameters, we could use any of the scalar Equations **Error! Reference source not found.**, **Error! Reference source not found.**, or **Error! Reference source not found.** in Section **Error! Reference source not found.** to estimate the extinction mixing ratio at each point. But to infer an extinction mixing ratio simultaneously consistent with all three measured variables, we instead use the calculated multivariate distributions illustrated in **Error! Reference source not found.**. Any value of the extinction mixing ratio is associated one-to-one with a backscatter mixing ratio using Eq. (20) and with a multivariate distribution given by a vector mean and covariance matrix as described in Eq. (30)-(32). A given measurement will be consistent with a range of overlapping multi-normal distributions, but by minimizing the Mahalanobis distance as a function of mixing ratio, we choose the distribution that is the best match to a given measurement. The Mahalanobis distance (Mahalanobis, 1936), discussed in detail by Burton et al. (2012), is a generalized unitless metric that describes the "distance" between a measurement point and a multivariate normal distribution. This calculation, choosing the extinction mixing ratio that best fits a given observation, is exactly analogous to the aerosol classification methodology describe by Burton

et al. (2012). However, instead of eight aerosol types described by multi-normal distributions, here we have a continuum of multi-normal distributions which sample the range of possible extinction mixing ratios from 0% to 100%. We choose the single multi-normal distribution which minimizes the Mahalanobis distance metric and therefore maximizes the probability of the measurement being consistent with the distribution. **Error! Reference source not found.** shows a time-height cross-section of the inferred extinction mixing ratio, f_{532}, for this flight.

Comment by T. Nishizawa

You proposed and demonstrated a novel method to separate two specific aerosol types (e.g., pollution+dust, dust+marine, or smoke+marine) using air-borne HSRL. Then I wonder why you focus on separating only two aerosol types. I guess that your method can be easily expanded to separate more aerosol type (e.g. three types, four types and so on) simultaneously depending on number of observed parameters.

It's true that the theory can easily be extended to more than two types. In the revised manuscript we have added additional theoretical development to show how it can be extended. For the data section, we focused on two types because examples with two types are very clear and easy to visualize and therefore provide useful illustrations of the technique. The application of the theory to the separation of three types in real data is also possible if the types occupy sufficiently distinct regions in the space defined by the measured intensive variables. For example, a case with dust (large depolarization), smoke (small depolarization, large lidar ratio and backscatter color ratio) and marine (small depolarization, small lidar ratio and backscatter color ratio) would be a good candidate case study. We will work on the separation of a three-component mixture as part of ongoing studies.