The authors would like to thank the reviewers for their comments and for this very useful input. Responses to comments are provided below, and appropriate changes have been made to the revised manuscript.

Reviewer #1, Ryan Moffet

This paper presents the first application of field measurements to the recently introduced entropy measures for quantifying aerosol mixing state. This paper builds on recent work by the authors to develop quantitative methods for single particle mass spectrometry. To my delight, I find this work to be excellent and I highly recommend its publication. I have listed a few minor suggestions below relating to the interpretation of the results.

Specific comments:

Section 2.2, Page 6: It should be stated what clustering algorithm is used to parse the data (K-means?). How were 10 clusters arrived at? Was it fixed in the algorithm? Also, why were other clusters not considered for the derivation of the RSFs? In general, it is not clear how or why clustering was used in this study and I think it would be good to briefly state this. What percentage of particles were represented in the 10 clusters?

This discussion of clustering actually referred to a previous article (Healy et al., 2013). In the current work, clustering was not performed, and instead each single particle was analysed directly. This work thus represents an evolution of the approach described in Healy et al (2013) in that the mass fraction calculations are applied at the single particle level, rather than being applied to 10 discrete clusters. To clarify this point, these sentences have been rewritten in the revised manuscript as follows:

“Approximately 1.75 million single particle mass spectra were collected during the MEGAPOLI winter campaign, and imported into ENCHILADA (Gross et al., 2010) for further analysis. In this work, an adaptation of the quantitative approach described by Healy et al. (2013) has been employed. Healy et al. (2013) previously derived campaign-averaged ATOFMS relative sensitivity factors (RSFs) for organic aerosol, elemental carbon (EC), ammonium, nitrate, sulphate and potassium using a combination of mass spectral marker ion intensities and concurrent bulk aerosol composition measurements. Healy et al (2013) then applied these RSFs to 10 discrete particle “types” assumed to have static chemical composition.

In this work, RSFs have been calculated at hourly resolution, and applied at the single particle level instead, in an effort to reduce the uncertainty associated with single particle chemical speciation estimates. Hourly average black carbon (BC) mass concentrations were measured using the MAAP and hourly average mass concentrations for ammonium, nitrate, sulphate and organic aerosol (OA) were measured using the HR-ToF-AMS. These data were used to constrain the ATOFMS RSF values as described in the Supplement. The five species chosen represent >90% of the composition of $PM_{2.5}$ measured at the site by mass (Bressi et al., 2013). The average contribution of potassium, sodium and chloride to the measured $PM_{2.5}$ mass concentration was relatively low (~3%) (Bressi et al., 2013), and therefore these species have not been included in the quantification procedure or in the diversity analysis here. The ATOFMS marker ions used for OA, BC, ammonium, nitrate and sulphate are the same as those employed previously (Healy et al., 2013).”

Figure S1: How do the RSFs in figure S1 differ from those presented in Table S1 of Healy et al. 2013. They appear to be quite different.

The RSF values reported in this work differ from those reported in Healy et al (2013) for three reasons. Firstly, in Healy et al (2013), each RSF value was expressed relative to that observed for potassium. However, potassium was not quantified in this work, and thus each RSF value is expressed relative to that observed for BC instead. Secondly, a single set of campaign averaged RSF values were used in Healy et al (2013), while in this work RSF values were calculated for every hour of the campaign in an effort to minimise the uncertainty introduced by composition-dependent matrix effects. Thirdly, the normalisation procedures used to calculate the relative peak areas (RPAs) of the marker ions are also different. In Healy et al (2013), a single normalisation was applied to each dual ion mass spectrum. In this work, the positive and negative ion mass spectra were instead normalised separately, in an attempt to minimise the uncertainty associated with the shot-to-shot variability of laser-particle interactions (Gross et al., 2000). The diversity and mixing state results obtained using combined and separate mass spectral normalisation were observed to be highly similar, however, as discussed in the manuscript. The RSF values obtained using each normalisation procedure, expressed relative to the RSF for BC, are compared to those reported in Healy et al (2013) in the following table:

Table 1: Relative sensitivity factors (RSFs) derived by constraining ATOFMS mass spectral relative peak areas using concurrent HR-ToF-AMS and MAAP measurements. RSFs for SO_4, NO_3, OA and NH_4 are expressed relative to that observed for BC in each case.

	BC	SO_4	NO_3	OA	NH_4
Healy et al 2013, campaign averaged RSFs, combined normalisation	1.00	1.86	2.00	9.79	15.72
This work, mean of hourly RSFs (N=610 hours), combined normalisation	1.00	1.54	1.69	13.07	21.19
This work, mean of hourly RSFs (N=610 hours), separate normalisation	1.00	0.88	0.96	4.33	7.06

Page 8, Density assumption for number scaling: I would think that a more accurate estimation of density could be arrived at using the mass fractions. Why or why wasn't this included?

While knowledge of the density of each single particle would be advantageous for achieving greater accuracy in the scaling procedures, this estimation was not attempted here for three reasons. Firstly, the density of the bulk aerosol was found to be relatively stable throughout the campaign (1.49±0.07 g cm^{-3}) (1σ) (Healy et al., 2012). Secondly, coarse particles that are expected to exhibit much higher material densities than 1.5 g cm^{-3}, for example crustal dust and sea salt particles, were found to have minimal contributions to the total scaled particle number (<2%). Thirdly, almost all particles were identified as internal mixtures of at least 2 species; BC and OA for example; or BC, OA and ammonium sulphate, for example. The effective density of BC particles can vary significantly depending upon shape, void space and coating thickness, while the effective density of pure OA can also vary significantly depending upon extent of oxidation. Thus, applying material density values and assuming shape factors is unlikely to be reliable in this case, for BC in particular, in the absence of supporting light scattering data (Moffet and Prather, 2009). Thus, a single density value was chosen for the conversion of aerodynamic diameter to estimated mobility diameter and the conversion of volume concentrations to mass concentrations. However, for sites impacted significantly by crustal material, or when light where single particle light scattering data are available, single particle density values should be used when possible.

Results, Pg 15: How might the exclusion of aerosol water affect the interpretation of the data from a modeler's point of view? Water is a very important component of aerosol, and one *would expect for a physically correct model it should be included. Perhaps this could be briefly discussed. Are there ways of measuring the water content of individual particles?*

This is a very good point. Particles were not dried prior to detection by ATOFMS, and therefore at least some liquid water may be present. The presence of particle phase water has been demonstrated previously to suppress negative ion sensitivity for desorption/ionization-based single particle mass spectrometers (Neubauer et al., 1998). However, particle phase water present in single particles sampled using aerodynamic lens assemblies similar to that employed here has been demonstrated to mostly evaporate, resulting in particle velocities in the sizing region similar to those observed for the equivalent dry particles (Zelenyuk et al., 2006), and thus also minimising water-related matrix effects. The presence of water on the ambient particles prior to sampling through the lens is thus not expected to significantly affect sizing accuracy or the relative sensitivities for the marker ions associated with each species in this work. If some particle phase water did remain however, the calculated ratios of the chemical species relative to each other would remain the same, but the mass closure approach employed here would result in overestimation of the absolute mass for each species present in each particle due to the absence of water in the calculations. Encouragingly, reconstructed mass concentrations derived from the single particle data agree well with the bulk aerosol instruments, which sampled dried ambient aerosol, for each species (Fig. S4). Nevertheless, drying particles prior to sampling would be prudent in future single particle mass closure studies.

Results, Pg 12: It is stated that D_i is around 2. What does this mean physically? That the population is typically only composed of two species on average? This might be expected for cation/anion pairs, but I would think that OC or BC would always be associated as well.

Wouldn't that make the D_i closer to 3? Or, perhaps this can be explained by the fact that OC is more prevalent in particles than inorganic species? If the values for D_i are compared to D_gamma (single particle diversity, usually always above 3) I think it starts to make more sense. I enjoyed thinking about this.

D_i represents the effective number of species present in single particle i. Physically, a pure BC particle thus has a D_i value of 1, while a 50% BC/50% OA particle has a D_i value of 2. D_i is approximately 2 for the most numerous particles because these are combustion-related internal mixtures of OA and BC with minimal inorganic ion content. Cation-anion pairs such as ammonium nitrate would also exhibit D_i values close to 2, but OA/BC internal mixtures are far more numerous in this dataset. D_i values closer to 3 are indeed observed for OA/BC internal mixtures that also contain inorganic ions (for example Fig. 4, middle panel). D_α represents the average single particle diversity, while D_γ represents the bulk aerosol diversity. The advantage of relating these terms is apparent in the following example: If 50% of the particle population were OA/BC internal mixtures with D_i ~2 and the other 50% of the population were ammonium nitrate particles with D_i ~2, then D_α would be ~2, but D_γ would be ~4. Relating D_α and D_γ thus reveals that this particle population is not comprised of a perfect internal mixtures of all 4 species, and that instead more than one type of particle is present.

Results, Page 16, Figure 7: This could probably go without mentioning, but I think it would be good to state if/how this figure was generated from the data. Section 3.4: I suggest moving section 3.4 (before current section 3.1) to the front of the paper before presenting the campaign averaged data. This way the reader fully understands what factors are controlling the average trends.

The following line has been added to the manuscript:

“The relationship between the average D_γ and average D_α values sorted by hour of the day (N=26 days), is shown in Fig. 7.”

The caption for Fig. 7 has also been rewritten:

“Fig. 7. The relationship between average single particle diversity (D_α) and average bulk population diversity (D_γ) values sorted by hour of the day (N=26 days).”

After rearranging the ordering of the results and discussion subsections, describing the overarching meteorological trends first was unfortunately found to detract from the flow of introducing the diversity terms through example cases. Thus the original subsection ordering has been retained in the revised manuscript.

Page 16. The sentence “Local sources contributed most significantly to aerosol mass concentrations under marine air mass conditions from 28/01/2010-07/02/2010” is potentially misleading considering the coarse mode was not measured. For these periods what was the PM2.5/PM10 split?

The average $PM_{2.5}/PM_{10}$ ratio, estimated from the combined TDMPS/APS distributions was 0.98 for the entire campaign, and for the period of interest was also 0.98, indicating that coarse mode mass contributions were minimal, even when the site was impacted predominantly by marine air masses. This is consistent with long-term off-line analyses of $PM_{2.5}$ collected in Paris, demonstrating that sea salt mass contributions are on average very low (~3%) (Bressi et al., 2013).

Conclusions, Page 17. The sentence “Aerosol mass spectrometry measurements have been used for the first time: : :” should be modified to state that Single Particle Mass Spectrometry and AMS measurements were used. It is widely understood in the community that AMS refers to the aerodyne AMS, and I think it should be clear that the ATOFMS was used here.

This has now been clarified in the revised manuscript.

Conclusions: I think it is important to state why the models are undertaking this task. Is it to simplify mixing state for larger scale (perhaps global?) models? One drawback I see with this approach is that the type of species is not explicit in the parameterization. How can one relate mixing state to atmospheric effect (e.g. direct radiative forcing) if the components cannot be "tagged" with composition. Perhaps some discussion of this is warranted.

The aim when applying diversity and mixing state metrics is not to simplify mixing state in existing models. Most state-of-the-art regional and global models do now consider aerosol chemical mixing state, at least to some extent, and thus a mixing state metric is expected to be particularly useful for constraining or validating these models. The mixing state metric will be useful for performing model intercomparisons in order to assess how differences between models depend on mixing state. In particular, the extent to which differences between model results and observations depend on aerosol mixing state can also now be assessed.

Regarding the loss of information, when particles are tagged with a diversity value, the compositional information is retained, both in single particle datasets and single particle resolved models. It is true that the diversity value does not depend upon *which* of the five species are present in the case of the approach taken in this work, only the extent to which they are mixed. The description of quantitative diversity and mixing state measures presented in this paper implicitly assumes that “species” should mean “chemical species”. Many other possibilities exist, however, including using species subsets (e.g., BC versus non-BC particles, or hydrophilic versus hydrophobic), and these alternate species definitions may be advantageous to relate mixing state to aerosol climate impacts (e.g. direct radiative forcing).

The conclusions have now been rewritten as follows to incorporate this discussion:

“The approach described here represents a new means by which to compare and contrast aerosol chemical mixing state in various environments globally. Potential applications include investigating changes in the mixing state of aerosol species as a function of plume age. Ambient aerosol diversity measurements are also expected to be useful for constraining or validating models that resolve aerosol chemical mixing state, given that most state-of-the-art regional and global scale models now resolve mixing state to some extent. In particular, this metric will be useful for performing model intercomparisons to assess how differences between models depend on mixing state. It will also be useful to assess how differences between model results and observations depend on mixing state. The description of quantitative diversity and mixing state measures presented in this paper implicitly assumes that “species” should mean “chemical species”. Many other possibilities exist, however, including using species subsets (for example particles that do or do not contain BC), and these alternate species definitions may be advantageous to relate mixing state to aerosol climate impacts, for example direct radiative forcing. Regarding future measurements, comparisons of χ values derived using single particle mass spectrometry and off-line single particle analytical techniques will also be valuable.”

Reviewer #2

General remarks:

The analytical framework, data and results presented in the paper of Healy et al. are of importance for the atmospheric science community and especially for those dealing with aerosol mass spectrometry since a new approach is presented to investigate the mixing state and particle diversity from a theoretical perspective. Although the approach is quite complex, needs several data processing steps prior to its application (i.e. a scaling of the single particle counts and clustering), and relies on auxiliary data as that from the Aerosol Mass Spectrometer (AMS), its capabilities as analytical tool are promising based on the results presented. The single particle mixing state and diversity measurements offer insight into atmospheric ageing processes and help to identify aerosol particle sources and their role in air pollution and climate. The paper is nicely written and well-structured. The detailed repetition/description of the newly developed framework and the mentioned examples help the reader thought the complex topic. The application of the framework to actual data demonstrates the usefulness and capabilities of this analysis approach. Although the results obtained with the new analytical tool do not directly present many new scientific findings, the paper is well-worth being published in ACP after minor revisions. The manuscript will lay the basis for this analytical framework to be adapted by the scientific community.

Minor remarks:

1) Page 3975, line 22: The "Shannon entropy" should be defined briefly. It might be helpful for the reader to explain in simple words how it is different to the well-known "thermodynamic" entropy to prevent confusion. This, however, can also be done in section 2.3.

The following explanation has now been added to the revised manuscript in section 2.3:

"Information-theoretic Shannon entropy is a measure of the uncertainty associated with a random variable and is not to be confused with thermodynamic entropy. The Shannon entropy value associated with each single particle (H_i) can be related to the mass fraction of each species present in that particle as follows:"

2) Page 3977, line 1: What is the definition of "organic aerosol"? It is understandable from the context, but "the mass fractions of ... organic aerosol" sounds odd. Although it is consistent throughout the paper and used in Healy et. al., 2012, 2013, already, but shouldn't it be named "organic carbon" or "mass fraction of organic aerosol constituents" in this context?

Here we refer to the complex mixture of primary and secondary organic compounds present in the particle phase. Organic aerosol was chosen for consistency with the widely used aerosol mass spectrometer (AMS) operational term. Organic aerosol remains a more accurate term than organic carbon, at least in this work, because oxygen and hydrogen also contribute to the mass loading quantified by AMS instruments. The following line has been added to section 2.1:

"The HR-ToF-AMS measures mass concentrations of non-refractory aerosol species including ammonium, nitrate, sulphate and organic aerosol. Here the latter refers to the mixture of primary and secondary organic compounds present in the particle phase."

3) P3978, l4: Does the AMS and MAAP uncertainty refers to the measured mass?

These uncertainty values do refer to mass concentration measurements. This has now been specified in the revised manuscript.

4) P3978, l17ff: "mass concentrations used twice in the sentence.

5) P3978, l19ff: Complex sentence – maybe make two out of it?

This section has now been rewritten as described earlier in response to Reviewer #1.

6) P3979,l6ff: What are the actual mass contributions of potassium, sodium and chloride, and where do you make a cut to not expect a significant impact on particle diversity? Here, I see a weakness of the paper as it is not clear how the addition (or even subtraction) of another chemical species – even with a small mass fraction – would affect the results. How would the model work for a more complex aerosol composition? Also, the uncertainty associated with the ATOFMS-derived particle speciation, and how this affects the diversity results should be moved and discussed in the main text, not in the supplement. These are important facts to understand the potential of the method, to judge its accuracy and to get an impression on the "natural" variability of the diversity and mixing state.

As discussed earlier in response to Reviewer #1, the mass contributions of these three species to $PM_{2.5}$ are low (~3%) (Bressi et al., 2013), and the mass contributions of sodium and chloride in particular are expected to be even lower for the size range investigated here ($PM_{0.15-1}$). However, it is certainly important to consider the effect that addition or removal of chemical species will have on aerosol diversity and mixing state. At the single particle level, pure NaCl particles would exhibit a D_i of 1.95, and if chloride were completely displaced by nitrate, pure $NaNO_3$ particles would exhibit a D_i of 1.79. Thus, sea salt particles have relatively low D_i values. The relative mass contribution of sea salt particles is so low, however (<3%), that the impact on average single particle diversity (D_α), bulk aerosol diversity (D_γ) and therefore the resulting mixing state index (χ), would be minimal. The contribution of trace elements that are internally mixed and not considered here, for example potassium present in biomass burning particles, is also minimal because the influence of species with low mass fractions are down-weighted in the Shannon entropy calculations. For example, a biomass burning particle consisting of 49.5% OA, 49.5% BC and 1% potassium will have a D_i value of 2.1 if the potassium content is considered, and will have a D_i value of 2.0 if potassium content is not considered. Therefore trace elements are not expected to impact the diversity and mixing state results significantly if excluded in this case study. However, the effect may be more significant in environments with higher contributions of crustal dust or sea salt to total aerosol mass concentrations.

Regarding uncertainty, there is undoubtedly high uncertainty associated with the scaling procedures employed here to estimate single particle composition. Assessing the absolute uncertainty at the single particle level is unfortunately not possible in the absence of knowledge of the exact composition of every particle. Thus, two approaches are investigated to estimate uncertainty. The first is how well bulk aerosol mass concentrations are reproduced when all single particles are considered. The agreement between reconstructed and measured bulk aerosol mass concentrations is within 30% for all five species (Fig. S3 & S4), although this approach is somewhat circular in that the bulk measurements are used to derive the relative sensitivity factors (RSFs) in the first place. The second approach; examining how the ATOFMS RSFs change as a function of time, likely represents the best method available to assess uncertainty in the ATOFMS measurements. Relative sensitivities for each chemical species will vary as a result of composition-dependent matrix effects and differences in desorption/ionization efficiencies for particles with different compositions. Thus examining the variability in the RSF values results in a higher uncertainty value for each species, but better reflects the extent to which the ATOFMS data are constrained by the support HR-ToF-AMS and MAAP measurement data. The uncertainty (1σ) is highest for ammonium (76%), which is sensible because this is the species for which the ATOFMS exhibits the poorest sensitivity (Gross et al., 2000). Uncertainty for the other four species is in the range 40-45%. However, it is important to note that the actual uncertainty at the single particle level may be higher.

The discussion of relative sensitivity factors and comparison of ATOFMS reconstructed mass concentrations with bulk aerosol measurements is discussed at length in the Supplement. The method described in this work is an evolution of an existing method, already described in the main text of Healy et al. (2013) in detail, and thus does not necessarily represent a new approach. The discussion of differences to Healy et al (2013), which is lengthy (>2000 words, 5 figures), was therefore moved to the Supplement to improve the flow of the main text, and to focus the narrative on introducing the diversity and mixing state concepts instead. This point is also discussed further in response to comment 8, and new additions to the main manuscript are described therein.

7) P3986,l27: Why does the ATOFMS not detect cooking-related particles? Explain briefly.

The absence of cooking particles in ATOFMS datasets has been noted in recent field campaigns where simultaneous AMS PMF analyses have identified significant contributions of cooking activities to total organic aerosol mass concentrations (Dall'Osto and Harrison, 2012; Dall'Osto et al., 2013; Healy et al., 2013). In each of these cases, clustering analyses of ATOFMS datasets did not resolve a particle type that exhibited similar temporality to the AMS cooking organic aerosol factor. ATOFMS single particle types related to vehicular traffic and domestic wood combustion sources were identified in these cases, however, and exhibited temporality that agreed well with their counterpart AMS PMF organic aerosol factors. One possibility is that the ATOFMS is not sensitive to primary organic aerosol associated with cooking activities, due to inefficient absorption of the desorption/ionization laser light (266 nm). Poor absorption efficiency at 266 nm has been reported for long chain aliphatic organic molecules likely to be present in cooking organic aerosol (Silva and Prather, 2000). The absence of internally mixed black carbon, which absorbs at 266 nm efficiently, and thus increasing the likelihood that particles will be successfully desorbed/ionized, may also hinder detection. The following lines have been added to the revised manuscript:

"Cooking-related particles are expected to have low D_i values because they are composed predominantly of OA, however the ATOFMS does not detect these particles efficiently (Healy et al., 2013), and therefore their number contribution is expected to be significantly under-represented in the ATOFMS dataset. This may be either due to an absence of internally mixed BC or poor absorption efficiency at 266 nm for cooking-related aliphatic organics (Silva and Prather, 2000)."

8) P3987,l2ff: Compare to comment 6 and discussion above; Here, the increase in χ can't be related to either the uncertainty of the method (due to the ATOFMS matrix effects) or the actual change in particle mixing state. Again, a number on the uncertainty of the method would be helpful.

The authors agree that the high uncertainty associated with the single particle composition calculations cannot be overstated. The single particle mass fraction calculations reported here represent estimated composition only. The RSF uncertainties calculated for each species have now been included in the manuscript as follows:

“A detailed discussion of the hourly RSF values obtained, the marker ions chosen and estimates of uncertainty associated with ATOFMS-derived particle speciation are provided in the Supplement. Briefly, the uncertainty associated with hourly mass fraction estimates for BC, OA, ammonium, sulphate and nitrate is 40%, 44%, 76%, 41% and 41%, respectively. The highest uncertainty is associated with ammonium, the species for which the ATOFMS also exhibits the poorest sensitivity. It is important to note, however, that uncertainty at the single particle level may be higher, but in the absence of accurate single particle composition measurements for comparison, this cannot be assessed for the ambient dataset reported here.”

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