Responds to referee comments

1. Introduction Line 25 There are I think two primary seasons of dust transport to the Atlantic, although one of these is at high level and has rather little impact on Cape Verde. Nevertheless it would be useful to clarify this. Also generally the utility of considering mass concentration of aerosol may be limited. The authors do not present sea salt data, but it will inevitably be present at relatively high concentrations. Hence unless the authors provide sea salt data to demonstrate that aerosol mass is related to dust loading, I would suggest eliminating the discussion of mass loading.

The line has been changed and now reads;
"The outbreaks observed at CVAO have seasonal patterns with peaks between December and March. However, dust transport to the Atlantic usually has two primary seasons which are in winter and summer but the transport during summer occurs at higher altitudes and is not usually observed at the CVAO. Such seasonal trends were reported by Chiapello et al. (1997, 1999, 2005) at Sal (a neighboring) island. The other months are mostly dominated by remote conditions".

As suggested, the discussion of mass loadings has been eliminated.

2.2 Methods XRF analysis is not widely used for aerosol analysis so I would suggest that the authors clarify a few details of the methods. Firstly explain about the "spots on the foils". Then explain "spots were placed on substrates" and why acid was added.

The particle spots are created due to aerosol particle impaction on foils surface during sampling. Depending on the number of visible particle spots on the polycarbonate (nuclepore) foils on each impaction stage, a given amount of visible particle spots are cut off from the foils and analyzed. The cut foil containing the visible spots is placed on a TXRF sample substrate holder and acid is added to help dissolve bigger particles on spots, improve the particle spot homogeneity and adhesion of the foil to the substrates surface.

Details have been included on manuscript on page 6 lines 10-13 and on pages 6 lines 16-17

As the authors note the "spots" seem to be the same size as the XRF beam, so confirmation that the whole of the sample is analysed is required. What reference materials beside the internal Ga standard were analysed?

Thank you for the correction. The right word is the "foil size" and not the spot size.

To verify that the whole sample was analyzed, a multi-element standard (C.P.A. Ltd, Lot N: N15521) including the elements Al, As, B, Ba, Be, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mo, Ni, Pb, Sb, Se, Sr, Ti, Tl, V and Zn was measured. This was done by dispensing 5 µl of the multi-element standard solution of known concentration on a TXRF substrate holder. The size of the 5 µl standard on the substrate holder is similar to that of the cut nuclepore foil containing the particle spots. The multi-element standard was then measured at three different angles as done with the samples. The obtained results were in good agreement (with a deviation of about ± 5%) with the values provided by the manufacturer thereby justifying this procedure.

A comment has been added on page 6 lines 27- 30 and Page 7 lines 1-2.

Line 24 what does "were not probed" mean, I assume the blanks were deployed with no air sucked through them, but the text is ambiguous.

The reviewer is correct, this sentence has been rephrased and the lines now read Page 7, lines 19-20; "The field blanks were filters that we deployed to the field and to the instrument but air was not sucked through them."

2.3 Back trajectories. The authors should explain rationale for the complex procedure they have undertaken to create 648 trajectories to describe each sample. This approach no doubt provides a very good characterisation of the air parcel's possible transport, but why was this done and does it yield a much better estimate than using just a few trajectories?.

As correctly said, it was used to provide an as good as possible characterization of the air parcels, which cannot be obtained as such using single back trajectories. Using this method, especially all the mixed scenarios can be much better classified compared to single averaged trajectories, making interpretation of results easier. Even though the method might seem rather complex, it is actually as easy to implement for us as calculating just a few single trajectories. The "ensemble" mode is an option in the PC version of the HYSPLIT model, which can just be activated instead of the "single trajectory" mode and the hourly starting times within a sample interval are fed to the model by a script. Thus, for us it basically makes no difference in the effort to calculate the 648 trajectories per sample.

I did not really understand the way that this information was merged with geographical data. The text suggests that landscape types had different colours - are these the colours in Fig 3?

This part of the manuscript was actually a bit misleading. Beyond just calculating trajectory ensembles and plotting them on a map, the trajectory method referenced in the manuscript (van Pinxteren et al., 2010) is in principle able to intersect the trajectory data with satellite-derived data on land cover classes in. This GIS approach yields quantitative indices for every sample describing the impact a certain land cover class potentially has on the sample. However, within this work, this analysis has not been done. We have only calculated the trajectory ensembles and plotted them on maps, where different colors represent different land cover classes (as described in the caption of Fig. 3). This aided the categorization of the samples into different scenarios.
We explain that in a hopefully more clear way in the revised manuscript. The part on the GIS analysis was removed for the reasons given above.

It seems inevitable that all samples at Cape Verde will have had a marine trajectory before arrival and the other classification into urban or agricultural areas seems at such a small scale that I cannot actually see it in Fig 3. Clearly a terrestrial versus exclusively marine trajectory is useful, as is differentiating between Europe and North America but, given the uncertainties in 4 day back-trajectories, I wonder about the value of subdividing landscape types beyond perhaps identifying the deserts. I note in the van Pinxteren et al reference cited this approach was used for urban source apportionment where I can readily understand it's utility, but it would be useful for the authors to indicate the way this high resolution source location improved their analysis for Cape Verde samples.

We agree with the reviewer that a subdivision of landscape types into too many (continental) categories does not make much sense in this study. This is actually the reason why we did not do the detailed GIS analysis as described above. We removed that part from the description of the trajectory calculation. In Fig. 3 the urban areas are not mentioned because indeed they cannot be seen at the scale of the map. The agricultural areas are the brown ones, which were misleadingly named as "continental" in the Figure caption. This has been corrected. Even though we do not use these land cover classes for a quantitative analysis, we would prefer to keep them in Fig. 3 as they allow for a better orientation on the maps.

3.1 Aerosol mass concentration. In the discussion of the mass concentrations and wind speed relationship for remote samples, it would be useful to know if the increase in mass at higher wind speed is simply related to higher sea salt concentrations.

Yes we see this effect mostly with sea salt concentrations from IC measurements.

3.2 Air mass analysis It is a rather pedantic point, but in the trajectory classes, even the SS, SU and EA classes must surely include an element of marine.

Yes they do, but the mass concentrations and main trace metal composition of these air masses differ thus we consider them as different classes. Information has been added on page 10 lines 22-24.

3.3 Trace metal concentrations I would suggest that having measured the concentrations in all the samples, the authors should include all the data including the "non remote" trajectory regions, even if they then choose to focus on the remote samples. The results can simply be tabulated. Their data base will be valuable to other workers.

We agree with the reviewer it will be more valuable to include the other measured data which may be useful for the scientific community. These data are available and will be published soon as a follow up paper to this.

Para 2: I accept the point that Marine North American air masses may be highly variable. This seems to me likely to reflect the air masses passing over high or low emission areas and/or at high or low altitudes. If the authors believe the explanation relates to wind speed as stated here, they should explain that further.

As the reviewer rightly confirms, the variability is link to the passage of air masses through different emission regions and altitudes, and due to high wind speeds this leads to mixture of different local air masses in an unpredictable manner, as in the case when wind speed would be low. A sentence has been included on page 12 lines 3-4.

The Weisel et al., 1984 paper does suggest quite large trace metals emissions from the oceans, but Hunter 1997 (in Sea Surface and Global Change eds Liss and Duce Cambridge Press) suggest a much weaker source. Much of the discussion in this section involves numeric comparisons to other data and I wonder if this might be simplified to a table. The general conclusion is that results are consistent with other data which is useful but could be said quite briefly.

The comparison section has been compressed and explained briefly.

Comparisons to some of the SEAREX Pacific sites should at least note that those are considerably more remote than Cape Verde from terrestrial sources. There is a concluding sentence "Generally metals from crustal origin: : :" which I disagree with. Clearly metals from crustal sources will be higher in European air masses, the transit times from source regions are shorter and the European air masses pass close to arid source areas.

This concluding sentence has been modified and the recommended aspects with respect to the transit times have been included in the concluding remarks on page 14 lines 3-5. Thanks.

However, I would suggest the authors statistically test if the Cu, Zn, Cr and Ni concentrations are higher in MNA air masses. I doubt from the data in Table 2 that this is true for Cr and Ni. It may be statistically true for Cu and Zn and if that is the case I think the authors might want to explore emission inventories to see if North American emissions are really high. The most recent compilation I know of (Pacyna 2001 Environ Rev 9 269-298) is of course from some years ago but it does not seem to suggest North American and European emissions are very different for these metals.

As recommended, corrections have been made with respect to Ni accordingly to clarify the discrepancies by removing Ni from this group. Concerning Cr we did not observe a great difference and thus the sentence has been modified to highlight only Zn and Cu differences since concentrations of Cr were almost similar in Europe and North America air masses. Page 14, lines 6-10

3.4 Size resolved analysis. I do not understand the point in this section about wind speed.

This aspect of wind speed is misleading here and has been removed from the revised manuscript. The intension was to explain the variability due to long range transport. This has been done on page 15 lines 2-3.

Also the authors make much of the fine mode crustal metals, but the size differentiation is essentially statistical so I am not surprised that a percentage of crustal associated metals are in the fine mode (SEREX saw this many years ago e.g. Arimoto et al 1989 In Chemical Oceanography vol 10 ed R Duce). Similarly metals such as Cr, Ni Cu, Zn and Pb will condense to existing aerosols after gas phase emission and this will give them a dominantly fine mode distribution, but it will not be exclusive as again SEAREX showed. In the case of Pb the massive decline in emissions probably means that re-suspension of contaminated soils may now be an important source.

These hints have been added and incorporated into the revised manuscript to substantiate the previous explanation on page 15 lines 17-22.

There is now evidence that marine colloids can be introduced to the atmosphere and could be a potential source of metals as the authors note, but in the absence of evidence such as from inter-element correlations the authors cannot demonstrate that this source is important compared to the well documented anthropogenic fine mode source. The importance of marine sources and their impact on aerosol Se are quite well documented (e.g. Arimoto et al., 1995 JGR 100, 1199-1213).

We do not claim the occurrence of these metals in the fine mode are solely from this source but only deduct from recent findings that these colloids could also be an additional source.
Now, this remark has been modified on page 15 lines 20-24 to read:
"The presence of such bioactive trace metals in the submicron particle range is usually attributed to their condensation on small particles after gas phase emissions from anthropogenic sources (Arimoto et al. 1989). However, recent findings also suggest that these metals may also have some oceanic origin coming from organometallic compounds in the ocean or from marine colloids…"

3.5 Enrichment Factors Ti is a perfectly suitable normalizing element, but I do not understand the comment "iron has a wider origin in oceanic regions than titanium".

The sentence has been clarified on page 16 lines 20 and now reads:

"Ti was used as marker for mineral dust."

The authors here assume an EF outside the range 0.7-2 represents enrichment. Given the variability in crustal composition others have set an EF of 10 as a threshold (e.g. Arimoto et al., 1989), so the authors may want to be cautious in interpreting relatively small EF values.

That is true. Actually EF's < 10 were few and have been considered to be similar to crustal origins since normalization is done with one reference material. A corresponding sentence has been added on page 16 lines 26-27.

The results here demonstrate that Fe has no enrichment and the authors might want to discuss this in relation to the argument that there is substantial emission of anthropogenic iron (e.g. Sholkovitz et al 2012 Geochim. Cosmochim. Acta 89, 173- 189). The results here seem to suggest that anthropogenic iron is minor.

We thank the reviewer for this point: We are aware of the existence of anthropogenic Fe but we did not observe Fe enrichment in our data, thus our suggestion that anthropogenic iron was not high in this region with respect to the air mass categories considered in this study. As explained before, the EF was evaluated from mean values making effects from events with significant enrichment averaged out. The variability of this data could be seen from the standard deviation of the reported mean values.
A corresponding sentence has been added on page 17 lines 25-29:

A minor point in the discussion of "group iii" I do not understand the units for a Ni emission factor of mg kg-1, kg of what? And in the next sentence, "fly ash" is not an "activity".

Kg of unleaded petrol and diesel

The sentence with fly ash and activities has been edited on page 18, lines 26-28 to read. "Cu and Zn are elements that are typical of emissions from anthropogenic activities such as fly ash, waste incineration and road traffic"

It would be useful to test conclusions from enrichment factors against emission inventories such as those of Pacyna.

Pacyna's emission inventory data have now been included in the conclusions. Page 19 lines 1-3.

The authors should perhaps note in the discussion of EF values that, for mixed source elements such as Mn, in dusty environments EF values will fall as the anthopogenic Mn is "diliuted" with crustal material.

We agree with the reviewer that for mixed source elements dilution may occur which might have also been the case for Mn. However, in this study we have not considered high dusty environments making us think dilution in the presented scenarios was not too strong.