**Reviewer #2:**

Reviewer 2 raises a number of issues in both the initial brief report and the later review published in ACPD. We thank the reviewer for the time spent on the manuscript and for the stimulus it has provided for us to improve the paper.

The initial brief report on our paper by this reviewer focused on three criticisms: 1) The text needed to be shorter, specifically the descriptive account of section 3 was not concise enough and should be cut by 20-30%; 2) Examples of modeling or back-trajectory analysis should be included; 3) We need to state our new findings more clearly. In response to this, for the ACPD version we reduced section 3 by 29%, added a new figure (3: back-trajectory analysis), and made changes to abstract, introduction and conclusions, attempting to emphasise the innovative nature of our database and the results.

The later review, to which we are now responding, has subdivided comments into 4 paragraphs. To ensure that we cover every point raised in these paragraphs, we have correspondingly further subdivided these into 1a-c, 2a-b, 3 and 4. Each point is discussed below:

**1a. "Describe more on what have been done on measurements of PM and other pollutants over the East Asian region".**

We are familiar with the extensive amount of work published on East Asian air quality, but for reasons of manuscript length chose to avoid a lengthy review of the subject. However, in response to the reviewers request we have extended the introduction:

"*In recent years a series of research articles has provided detailed information on these mixtures of natural and anthropogenic PM over East Asia, especially as a result of the 2001 Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) (e.g. Uno et al., 2003; Seinfeld et al., 2004; Zhang et al., 2004; Kim et al., 2005; Park et al., 2005 and references therein). Important contributions to the understanding of atmospheric PM chemistry in East Asia also include those using ACE-Asia data (e.g. Arimoto et al., 2004; Satake et al., 2004) as well as detailed observations on individual airborne particles (e.g. Zhang et al., 2003;2006; Kojima et al., 2006; Tobo et al., 2009) and on transboundary SIC (e.g. Nishikawa et al., 1991; Shimohara et al., 2001; Mori et al., 2003; Trochkine et al., 2003; Zhang et al., 2004; Uno et al., 2007; Aikawa et al., 2010; Fairlie et al., 2010; Kong et al., 2010). There are also papers specifically dealing the presence of anthropogenic metal-rich particles within Asian atmospheric aerosols (e.g. Kim et al., 2003; Arimoto et al., 2004; Park and Dam, 2010).*"

**1b. "Clearly describe the motivation of this study and focus on the difference between this study and previous work".**

To set the scene and clarify the difference between our study and that of previous published work, the above paragraph continues:

"*However there is still a relative paucity of detailed information published on the variations in chemical concentrations of the aerosols people are inhaling during these transboundary pollution events. Many chemical data provide only partial analyses of PM size fractions and/or analyse Total*

*Suspended Particulate Matter (e.g., Okuda et al., 2008; Yuan et al., 2008; Kim et al., 2009; Kong et al., 2010) rather than strictly the inhalable fraction (Cheng et al., 2005; Chan et al., 2008).*"

In the original text we explained the background to our study of transboundary air pollution events affecting Japan. Back in 2010 we had conducted a short pilot campaign to analyse 24-hour $PM_{10}$ filters collected during three transient transboundary pollution episodes crossing western Japan. The results were interesting, and have been published, but we felt the need to explore in much greater depth the arrival and impact of the transboundary intrusions. With this in mind, we designed a much more ambitious new campaign which involved collecting continuous chemical data on an hourly basis over 6 weeks in spring 2011. The current paper is the first and only one to present results from this extensive and unique new database.

We have provided more emphasis on the motivation of the study as requested (lines 80-112): "*Our results (from the pilot study) confirmed the pronounced bimodality and inhomogeneity between natural and anthropogenic PM in East Asian transboundary aerosol intrusions (Moreno et al., 2012). Furthermore, although the number of filters analysed in the pilot study was relatively small, it was nevertheless enough to demonstrate the highly metalliferous and chemically complex nature of sulphatic plumes arriving from industrial China. Such plumes cross Japan, creating regional pollution clouds that dissipate only slowly due to the dominance of atmospherically persistent submicron accumulation mode PM. It was this aspect of the chemical data, rather than the already well-characterised nature of the "natural" Gobi-Desert derived Kosa intrusions, which seemed to us in more urgent need of further study.*

*In this context this manuscript moves forward by presenting a new database collected during a 6-week period of continuous hourly and daily measurements in Kumamoto, SW Japan. The study adopts an unusually multi-analytical approach by integrating results from Particle Induced X-ray Emission (PIXE), Inductively Coupled Plasma Mass Spectrometry/Atomic Emission (ICPMS/AE) spectroscopy, chromatography and thermal-optical transmission methods, allowing comparison between hourly (Streaker) and 24-hour (filter) data. Such data are unprecedented in the chemical detail they offer on Japanese air chemistry during transboundary aerosol inflows. The Streaker data in particular provides over 1000 hourly measurements of major and trace elements for each of the coarse and fine size fractions, a very large number which allows us to perform a positive matrix factorization (PMF) analysis and so separate differently sourced components within the aerosol mixture.*

*The primary aim of this new campaign was thus to provide further insight into the complex atmospheric mixing processes and changes in air quality that occur within transboundary receptor areas lying immediately downstream from East Asian aerosol outflows. Using a time resolution measuring hourly as well as daily fluctuations allowed us to compare polluted and "clean" days, and to develop a clearer idea of the reality of what people are inhaling during these pollution episodes. This database would allow us to resolve in more detail than any previous study the speed, duration and chemistry of different types of pollution episodes reaching Japan. We consider these kind of previously unavailable data as prerequisite information for those tasked with provided health advice to affected populations prior to the predicted arrival of transboundary atmospheric pollutants.*"

**1c. "Draw a few scientifically sound conclusions which have not been identified by previous studies and can advance the community's knowledge on trans-boundary transport" and "Summarize the unique features of the air quality data measured in this study in the context of previous works".**

We have expanded the conclusions section and added five clear conclusions which summarise the main findings of the paper. The unique nature of the Streaker data, and the unprecedented number of full ICP analyses has been mentioned above. The main conclusions of the present study are summarized as follows:

*1: Based on the PMF analysis, the coarser PM fraction (PM2.5-10) is dominated by mineral dust and sea salt, whereas the finer fraction (PM0.1-2.5) is largely sulphate that includes As-bearing and Zn-Cu-bearing particles. The PMF results also indicate that metals can be incorporated into aged sea salt.*

*2: Mineral dust arrivals are highly transient, usually peaking over a few hours before rapidly declining. This contrasts with the more atmospherically persistent sulphatic transboundary aerosols, although these may decline in their toxic metal content as they age.*

*3: CFORS is an excellent predictor for transboundary sulphatic PM intrusions. However, our hourly monitoring detected a few CaAlSi mineral dust peaks that were not predicted by CFORS. These transient mineral dust peaks all occurred during major sulphatic events indicating an atmospherically persistent but poorly mixed sulphatic PM cloud containing areas rich in Al silicates.*

*4: Transboundary aerosol plumes in Eastern Asia are highly chemically inhomogeneous, depending on the geology of the source area (e.g. calcareous or not), the nature of the pollutants they incorporate during their generation (producing contrasting mixtures of metalliferous PM), and the amount of mixing during the varied physico-chemical conditions they encounter during transport (nitrate and sea salt content).*

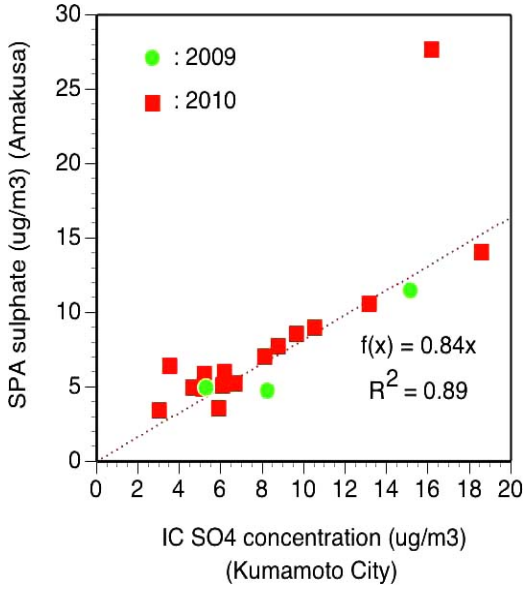
*5: The dominant influence on air quality at Kumamoto, and by analogy probably across much of the Japanese Archipelago, is the regular arrival of air masses contaminated with sulphate and toxic metals and sourced from mainland Asia. The contrast between atmospheric conditions under westerly and easterly airflows crossing Japan is striking and a cause for concern with regard to human health effects.*

**2a. "All data and analysis are based on only one site. I wonder if it is possible to compare the results of this study to other measurements (e.g., the EANET measurements)….. The measurements were made only in SW Japan and only lasted for a few weeks. I wonder how they could be used to infer the air quality over other more populated regions in Japan and other seasons and years……Show to what extent the air quality is impact by local sources."**

Indeed, the data in the manuscript were measured at just one site, but we are confident that the site is a good one for measuring what we were trying to investigate: namely transboundary PM intrusions into western Japan. However, we accept that we did not clarify our justification for this adequately and have added to the text in section 2 (Methodology, lines 122-148) to cover the main points given in detail below.

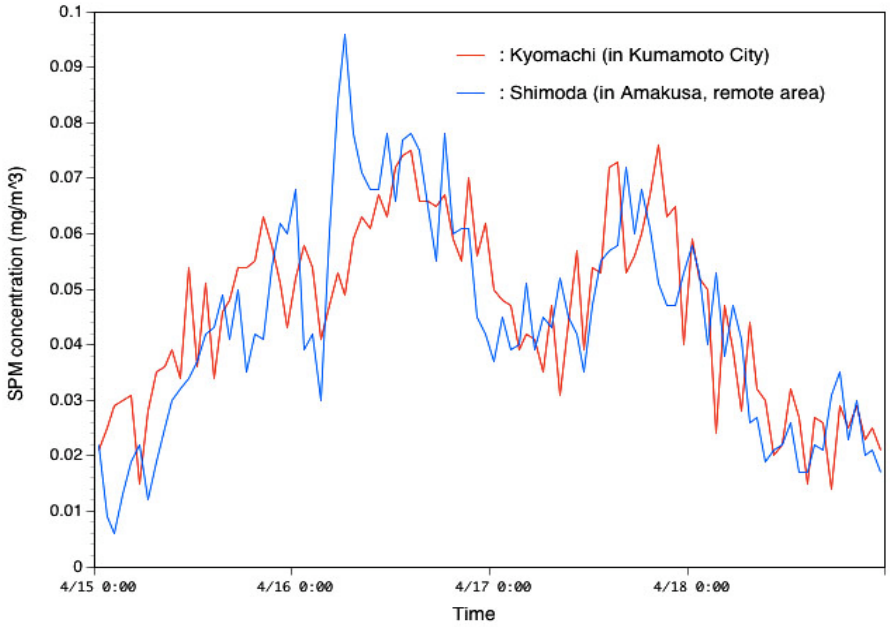
Firstly, we have access to air quality data from 33 monitoring stations (both urban/suburban and rural areas) provided by the Kumamoto prefectural government (http://taiki.pref.kumamoto.jp/kumamoto-taiki/index.htm). Having looked at the 2011 spring data our PM data from Kumamoto city are closely correlated with those measured at these different monitoring stations sited across the prefectural area, and our monitoring site can be classified as typical "urban background" for this city.

Secondly, we had previously "ground truth tested" our Kumamoto monitoring site by comparing $SO_4$ concentration obtained by ion chromatography (IC) of Kumamoto University filter samples with that by Sulfate Particle Analyzer (SPA) at Amakusa, a remote coastal on the western coast of Japan, 90km SW of Kumamoto (Nagatani et al., 2012). The figure below compares Kumamoto and Amakusa $SO_4$ data in 2009 and 2010:

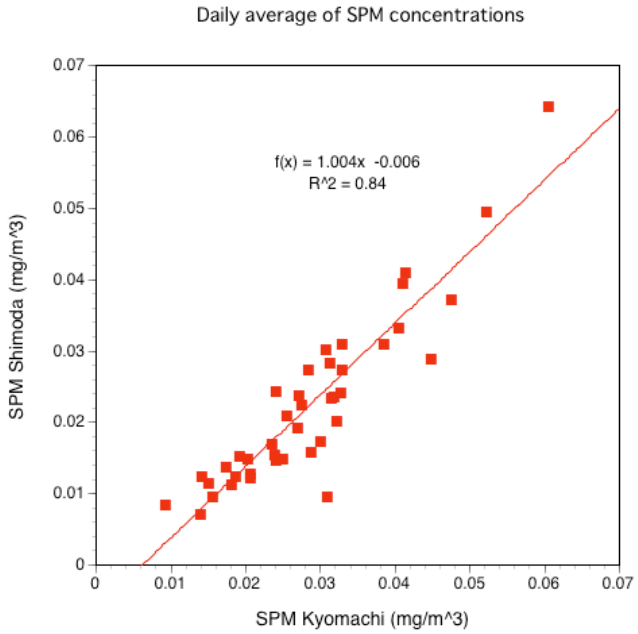



The data correlate well ($R^2$=0.89) except for one sample. The slope of the fitting line is 0.84. Because SPA is reported to yield ammonium sulfate concentration 20% lower than that obtained by IC filter analysis (Schwab et al., 2006), it seems that sulfate concentration in Kumamoto City was not significantly higher than those in Amakusa. Unfortunately in 2011, the SPA was not in good condition and sulfate data obtained at Kumamoto were consistently too low, although the instrument did record temporal variation of sulfate concentration and at least this correlated well with Amakusa.

Additional supporting evidence that aerosol concentration (mass of suspended particulate matter, SPM, <10μm diameter) in Kumamoto City was similar to that in Amakusa in 2011 April is provided by the figure below. The figure is based on the data monitored and publicized by the Kumamoto prefectural government (mentioned above). Kyomachi is the monitoring station nearest to Kumamoto University (where we collected our samples), whereas Shimoda is the one in Amakusa.




Similarly, the figure below shows daily average of SPM concentrations in Kyomachi (Kumamoto City) vs. Shimoda (Amakusa) during the campaign period (March 18 – April 27, 2011). As suggested by the figure above (time series of SPM concentrations in Kyomachi and Shimoda), the concentrations in the two locations correlate well with each other. The fitting line shows that SPM concentration in Shimoda is lower than that in Kyomachi by 0.006 mg/m$^3$. This appears to be the amount of "additional" local PM in Kumamoto City. With increasing amount of transboundary aerosols, the local influence would be less significant. Sometimes (on Kosa or pollution days) SPM concentration in Shimoda exceeded that in Kyomachi.




Furthermore, unpublished data on Optical Particle Counter data measured at Amakusa during the same period show time-series patterns are strikingly similar to those observed by us in Kumamoto during our campaign (Prof. D. Zhang, Kumamoto Prefectural University, *pers. comm.*).

Thirdly, a recent paper by Kaneyasu et al. (2011) compares $PM_{2.5}$ concentrations during 2009 and 2010 in Fukuoka (the largest city in Kyushu, 90km north of Kumamoto), Nagasaki (medium-sized city 75km west of Kumamoto), and Fukue Island (remote site 100km west of Nagasaki). It demonstrates that from March to May, $PM_{2.5}$ concentrations in Fukue Island were *higher than in Fukuoka* (Kaneyasu et al., 2011). So it is clear that air quality in Kyushu is affected more by transboundary aerosols than by local emissions, even in Fukuoka, a city far more highly populated and industrialized than Kumamoto.

**2b. "description on whether or not the situation of spring 2011 is more/less likely to transport dust and other aerosol pollutants from China."**

Eastward transport of dust and pollutants to Japan from Mainland Asia is a well-documented phenomenon, and the problem is most acute in springtime, when the vast majority of the classical Kosa intrusions have occurred. This is why we chose this epoch of the year for our sampling campaign.

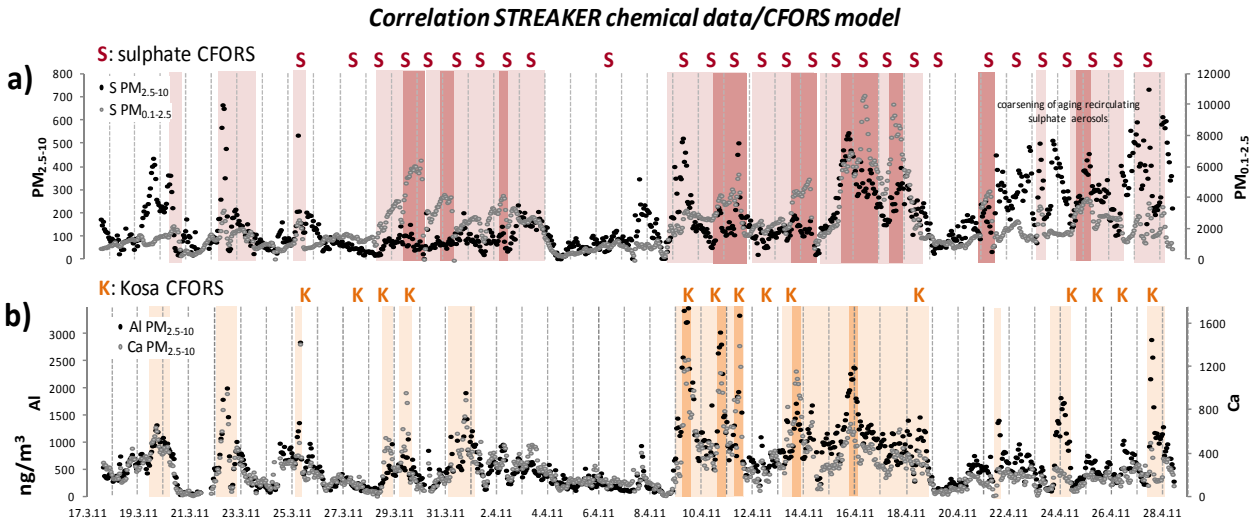
The Japan Meteorological Agency provides data on Kosa observations:
http://www.data.kishou.go.jp/obs-env/kosahp/kosa_table_1.html

These data record the number of the days when Kosa was observed somewhere in Japan during each month in each year since 1967. According to this, in spring 2011 there were fewer Kosa intrusions than in recent years.

**3." Should be directly compared and analyzed to model prediction by the Chemical Weather Forecasting System (CFORS). In particularly, I wonder how well CFORS captures these peaks for dust and sulphate aerosols."**

It was never our intention to attempt a detailed comparison between our dataset and the CFORS database, which we used as a guide as to when we might expect transboundary intrusions. However, in response to the Referee's request we have spent considerable time on this interesting aspect, and summarize our conclusions as follows:

At the start of our sampling campaign CFORS data were unavailable due to the disruption caused by the Tōhoku Earthquake on 11/03/11. The system was reinstated on 25 March, however, so that we were able to follow CFORS predictions for the remaining 34 days of the campaign. During this period there were only four days when CFORS $SO_4$ predictions failed to correspond with our chemical data collected on the ground. In all four cases sulphate pollution was predicted but not recorded specifically at that time, this being due either to slightly delayed arrival (27 March) or more rapid passing (11 and 19 April) of the sulphate cloud, or a "featheredge effect" when the predicted margin of the polluted airmass just missed the Kumamoto area (6 April). The figure below (a) superimposes on the Streaker data the days when $SO_4$ transboundary intrusions ("S") were forecast by CFORS (This is added as a new Figure 5 in the paper). We conclude that CFORS is in general an excellent predictor of transboundary sulphatic air masses.




In the case of transboundary "crustal" dust events our data demonstrate that not all silicate dust peaks can be attributed directly to Kosa events forecast by CFORS. During our sampling campaign CFORS predicted the presence of dust intrusions on 14 separate days (marked as K on the above figure b), and all of these were detectable on the ground as transient CaAl peaks in our Streaker dataset (with the one exception of the 27 March delayed arrival also noted for $SO_4$). There were, however, three episodes when transient CaAl peaks occurred despite the fact that no mineral dust event had been forecast. In all three cases these episodes occurred during the major sulphatic events. This observation indicates that the atmospherically persistent sulphatic PM cloud remains chemically heterogeneous, containing areas rich in Al silicates (presumably some combination of natural minerals and flyash), the passing of which are recorded as transient peaks by the Streaker data (for example see 31 March,16 and 21 April on Figs. 4 and 5. It would be interesting to know how much of the natural mineral dust incorporated within the sulphatic intrusions is derived from Kosa desert materials, and how much has originated from a more proximal source *en route* to Japan.

These results have been added to the conclusions (see lines 410 and onwards).

**4. "More improvements are needed in the conclusion section (i.e., section 4), particularly for policy implications. The measurements were made only in SW Japan and only lasted for a few weeks. I wonder how they could be used to infer the air quality over other more populated regions in Japan and other seasons and years."**

As we have explained above, we can present strong evidence to support our rooftop monitoring site at Kumamoto as being suitable for recording the impact of transboundary PM intrusions into western Japan. As the CFORS data demonstrate, these transboundary intrusions commonly move further east to cover much of the Japanese archipelago. Indeed these events are so common, especially in springtime, that researchers have described transboundary air pollution in Japan as "quasi-permanent" (Lasserre et al., 2008).

The campaign lasted for 6 weeks, covering much of the Japanese springtime for that year. We would argue that the data can be considered as broadly representative of springtime atmospheric

conditions in Western Japan, but cannot be used directly to infer air quality in other more populated parts of Japan. It is likely, however, that the patterns of sulphatic pollution evidenced during our campaign occur on a regional level, and we would expect the rise and fall of pollutant levels in response to these transboundary PM intrusions to be similar elsewhere across the country, although becoming progressively diluted eastwards.

We have expanded the Discussion and Conclusions section, adding the 5 conclusion points mentioned above. With regard to political policy implications, we feel that this is beyond the scope of our paper.

Finally, the reviewer argues that our paper is **"just describing the time-series of measurements and is lack of new findings which significantly advance the community's knowledge on trans-boundary transport".**

We consider that one of the major findings in this study is how large the continental impact on Kumamoto air quality is. Our 6-week database allows us to demonstrate the impact of mainland Asia-sourced atmospheric emissions on Japanese air quality more convincingly and in greater detail than any previous study published so far, especially because our continuous record allows us to make comparisons between polluted and clean days. Also, to the best of our knowledge this is the first report of hourly trace metal data of atmospheric aerosols measured in Japan. The detail we provide on PM chemistry and levels during six continuous weeks of transboundary intrusions into Japan is unprecedented. The hourly rise and fall of the different aerosol components, especially the metalliferous sulphatic PM, has not been demonstrated with such graphical clarity. Having obtained over 8500 continuous hourly measurements using PIXE analysis of the Streaker data, we were able confidently to perform PMF source apportionment analysis: again to our knowledge this is the first time this has been done on such a large dataset of transboundary Asian aerosols. Figure 4 is new, innovative and graphically illustrates the daily reality of springtime air pollution impacts in western Japan. Similarly, again to the best of our knowledge, the number of full ICPMS analyses (37 samples) presented in the paper is far higher than any paper previously published in English on Japanese aerosols.

The fact that the data were collected from just one site does not detract unduly from the study, not only because (as we have shown above) our monitoring site is not seriously affected by local emissions, but also because CFORS shows quite clearly how transboundary sulphatic intrusions are regularly spread throughout Japan. Kumamoto in Western Kyushu is situated in one of the worst areas to be impacted (which is why it was chosen), but it is far from being the only Japanese city seriously affected by this problem. The database allows us to resolve in more detail than any previous study the speed, duration and chemistry of different types of air pollution episodes reaching Japan. We consider such information as especially useful to health care providers advising affected populations prior to the predicted arrival of atmospheric pollutants.

References:

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