Response to the reviewers' comments (2) on HESS-2009-180

Dear Prof. Alcala,

Your detailed comments and suggestion are appreciated. Below are our responses. Look forward to your further advice.

Sincerely,

Huade Guan
On behalf of the co-authors.

General comments

The manuscript presents the correlation analysis as tool for estimating spatial atmospheric bulk chloride deposition (BCD) in east of Adelaide, South Australia. The BCD data are needed for spatial groundwater recharge estimation through the chloride mass balance (CMB) method. They are few successful experiences on the CMB method application in hilly coastal areas. In these areas, the high spatial gradients of BCD can constraint accurate groundwater recharge estimation. Nevertheless, the statistical approach developed in this work not entirely improves the estimation that classically could be achieved by geostatistical methods, such as kriging with external drift, co-kriging, etc. Analyzing a well-known set of variables controlling the BCD in hilly coastal areas, no general rules are introduced. The role of dry deposition in BCD is not defined or quantified. Without a clear justification, some anomalous samples are removed in order to reduce uncertainty, artificially. This means a serious problem to justify the goodness of the correlation analysis. Uncertainty due to spatial errors and those derived from approaches are added to the natural variability needed to asses' long-term groundwater recharge variation due to land uses changes. Conceptually precipitation and their stable isotope signature can be modelled by quasi-linear relationships with temperature or similar potential induced covariates as elevation, but not the chloride content in rainfall (as authors propose), nor the wind-blown halite from marine aerosol and sea-breezes, urban and industrial activities and lithology, which are unknown in the study area. Section 2.1. A brief hydrogeological description is needed. Provide data on population density, industrial activities, etc., as well as other climatologic data as temperature, potential and actual evapotranspiration, soils, vegetation cover and lithology.

Discussion

First, about the mapping methods, the de-trended residual kriging approach (RK) presented in this manuscript provides an optional method for chloride mapping. The authors don't mean to exclusively replace other methods such as cokriging (CK) and kriging with external drift (KED). The three methods share that secondary variables are used to estimate spatial distribution of the primary variable. In general, they are not new. The problem is which secondary variables are appropriately used. With KED, the trend can be automatically searched when secondary variables are chosen. However, as Isaaks and Srivastava pointed out in their book (*An Introduction to Applied Geostatistics*, p532) that " though automatic methods exist for finding the trend that is best in a statistical sense, this trend may not have the support of common sense and good judgement". This problem is more important where the data points are sparse, such as the case in this study. Thus, we follow Isaaks and Srivastava's suggestion (p532) that "… it is wiser to choose a trend based on an understanding of the genesis of the phenomenon, subtract this trend from the observed sample values to obtain residuals …". The contribution of this study is to determine, based on the understanding and correlation analysis, which secondary variables and at what form are appropriate to estimate the trend. The regression we applied to search the trend is easier to include secondary variables other than (x and y), and still keep the method simple, in comparison to the other two. Actually, from our earlier comparison of this method and CK, the mapping result from our method appears better than cokriging with one secondary variable (Guan et al., *Journal of Hydrometeorology* 2005). The additional benefit from the regression is that we recover the beta coefficients to use for interpretation, e.g. we can discuss the effect of windward and leeward slope. One point we would like to mention is that the mapping exercise in this paper has a ratio of data points to mapping pixels of one 500^{th}, in contrast to earlier kriging mapping work with this ratio of about one 25^{th}, such as in Alcala et al. 2008.

That the two data points were removed from the correlation and regression analyses, is based on our existing understanding of atmospheric chloride transport and deposition, and the prevailing wind direction. The primary assumption we relied on to define the trend (not necessarily a function of x and y) is that the chloride comes from marine aerosols by wind and deposits on land by dry and wet deposition. The regression is designed to examine how the effect of these processes on BCD over the study area can be estimated. Thus, other processes, such as local chloride source other than the dominant marine chloride from the prevailing upwind direction, should be excluded. Because of the irregular coastal line, the two sample sites that were removed from the correlation/regression analyses) locates far downwind from the dominant chloride source to the study area, but with higher bulk chloride deposition due to closeness to the local sea and saline lake water (southeast corner of the study area). The chloride from these local sources does not contribute to chloride deposition in the whole study area, based on the wind direction data. Thus they are removed from the correlation/regression analyses. This treatment is also in line with Prof. Goovaerts' comments on about the stationary assumption of the regression coefficient. Ideally, we could do regression for different subregions to capture different related physical processes, such as regression applied for each topographic facet in PRISM model. But due to the sparse data points, this is not feasible in this study. We need to use one regression the capture the dominant processes impact on BCD.

About the dry deposition, our data (bulk chloride deposition only) do not allow us to separately analyse dry and wet deposition, although partial correlation analysis infers some possible processes influencing dry deposition differently from wet deposition. And separation of dry and wet deposition is not the focus of this paper. Nevertheless, we agree that to better understand BCD in the coastal area, it is important to separate wet and dry deposition.

We totally agree that "Uncertainty due to spatial errors and those derived from approaches are added to the natural variability (is) needed to asses' long-term groundwater recharge variation due to land uses changes.". Thus, we create the uncertainty map in addition to the chloride deposition map. We will improve this uncertainty map, following Prof. Goovaerts' advice.

In terms of simple regression model, we don't intend to estimate "the chloride content in rainfall". Our purpose is to estimate BCD. We show that our simple regression with two predictor variables (X and $\beta \sin\alpha$) capture about 70% spatial variability of BCD at 15 sites over the study area. This result suggests that BCD can be modelled with linear relationship over our study area probably because of one dominant marine atmospheric chloride source. However, this linear regression does not produce estimates close to the observation in the southeast corner of the area, due to probably a local chloride atmospheric source. This is why in ASOADeK model, we apply residual kriging to improve the final mapping estimates. In terms of elevation, our correlation result suggests that it is not significant in interpreting BCD.

The Adelaide and Mount Lofty Ranges area houses about 1.2 million population, with primary industry including health service, education, winery, and tourism. No significant air pollution sources of chlorine exist in the area. The lithology in the MLR is late Precambrian metamorphous sedimentary rocks composed of shale and sandstone, and minorly limestone (Preiss, 1987). Mean daily temperature over the area is about 15-18°C. The annual pan evaporation at a location of 600-mm precipitation (about area-average value) is about 1500 mm.

Actions
The above discussion is incorporated to the text wherever possible. A new uncertainty map is produced.

Specific comments

Page 5852
Line 6-9. This asseveration requires providing a tentative groundwater turnover time value. Line 10. Show generic applications. Chloride deposition maps are needed for,: : : This is a new methodology that improves : : :

Discussion and actions
The forest clearance impact on changing chloride equilibrium status, is a background and motivation for this study. Details in how we use the mapping result to examine the issue is presented in a separated paper. Apparently, this leads to some misunderstanding. This part is now removed.

Line 14. What means terrain aspect?.

Discussion and actions
It means slope orientation. It is now explained when the term appears first time.

Line 16-17. What type of gradient?, average in the catchment, from the coastline,

Discussion and actions
This is the gradient inferred from the regression, i.e., the regression coefficient of the x term. It is an average gradient in the study area. It is now explained when the value appears first time.

Line 20. Average uncertainty. What type?. Due to natural or inter-annual variability, as kriging uncertainty by spatial interpolation, due to simplifications in approaches to calculate BCD?. Define and comment: : :

Discussion and actions
We mean mapping uncertainty. We add an equation to clarify this issue.

P5853
Line 3. Describe accurately the CMB methodology for generic cases: A typical: : :

Discussion and actions
We agree. The part is re-written.

Line 6-7. Precipitation in the catchment?. May be effective precipitation to the land?.

Discussion and actions

It is precipitation to the catchment, not the effective precipitation (we assume you are talking about precipitation less interception loss). Precipitation brings chloride into the catchment no matter where it falls (either temporally on canopy or directly to the ground).

Line 7.Cg is not in groundwater. Theoretically, it is below the root zone and assumed to be equal in groundwater (Scanlon, 2000). Line 6-9. Improve these sentences. Define the CMB method for transient condition with possibility of chloride retention in the soil, and steady conditions for average recharge evaluations (in the sampling period o from yearly values). They are a problem of time-scale.

Discussion and actions

Apparently it is not clearly written here. CMB can be applied with groundwater chloride, or with soil water chloride, as long as they are in equilibrium with the local surface conditions. We rewrite this part.

Line 13-14. Some mixing cells numerical methods are based on groundwater and surface flow asseverations. See publications of Adar and Neuman (1988) Adar et al. (1988), Gieske and De Vries (1990).

Discussion and actions

We agree with you, and slightly rephrase the statement as follow. "The CMB method does not require knowledge of dynamic hydrological processes.(Although with such information, it would help to apply the CMB method more reliably)".

Line 14-15. Classically the CMB method was a very uncertain method in mountainous coastal areas (see Gasparini et al., in Canary Islands, Rosenthal in Israel, etc: : :).

Discussion and actions

We agree. One challenge to apply CMB in the coastal areas is the high spatial variability of chloride deposition. To estimate BCD spatial distribution is the motivation of this paper.

Page 5854
Line 2. Describe other possible not evaluated sources of chloride in deposition in coastal areas, such as lithological and anthropogenic, which can be relevant up to 30 % of bulk chloride in coastal plains and up to 50 % in summit coastal areas in polluted zones. See the use of the Cl/Br ratio for identifying sources of CBD in Spain (Alcalá and Custodio, 2008b).

Discussion and actions

Apparently, atmospheric chloride sources are quite site-specific. No obvious lithological and anthropogenic sources of chloride are observed or reported in this area. It would be good to examine whether additional sources add chloride to the study area if data is available. The logic we follow in this study is that, we assume that the dominant chloride deposition comes from the ocean by westerly wind and precipitation, which can be captured by the regression. If there are additional chloride sources, BCD would deviate much from the regression estimate, such as sites #16 and 17. The good performance of the regression with two predictor variables on BCD at the remaining 15 sites, supports (although not exclusively in terms of logic rule) that westerly marine chloride is the primary source for the study area. This is consistent with several pervious chloride deposition studies in this and other areas in

Australia. Nevertheless, we agree that the complexity of multiple chloride sources should be brought to attention.

Line 19-20. References about wind direction and intensity controlling CBD should be attached. Line 24-25. Minor et al. in Nevada (USA) or Carratalà et al. in Eastern Spain used similar geographical covariates for mapping CBD.

Discussion and actions
The prevailing wind direction effect on chloride deposition is obvious, under the assumptions that atmospheric chloride is primary from marine aerosol, through either wet or dry deposition. Thanks for providing the two references. We have Carratala et al paper. Similar to the two earlier mapping exercises that we cited, kriging is used in Carratal's paper. We could not locate Minor's paper on BCD, but found Minor's groundwater recharge paper on J. of Hydrogeology. Carratala's paper is now cited.

Page 5855
Line 5-7. Explain it better and reference.

Discussion and actions
In the coastal area, where BCD varies significantly over a few kilometres, the big limitation to create a good BCD map is the sparse observation points. We discuss this prior to line 5-7. If some influencing factors can be quantitatively related to BCD, it would be useful for BCD mapping. We re-write this part as follow.

Can we incorporate some associated physical process information, including coastal distance dependence, so as to make more reliable estimates for chloride deposition to form a basis for BCD mapping? In this context, geostatistical approaches, such as residual kriging (RK), kriging with external drift (KED), and cokriging (CK), can be used to incorporate secondary variable information in the mapping (Isaaks and Srivastava, 1989; Goovaerts, 2000; Guan et al., 2005). Because of the difficulty to obtain appropriate secondary variables and functions, RK is chosen, in which the secondary variable effect, often called trend estimate, is determined first (Isaaks and Srivastava, 1989) (p532). Similar approaches have been successfully applied in precipitation and rain water isotope mapping in mountainous terrains (Guan et al., 2005; Guan et al., 2009). The objective of this study is to examine the influencing factors associated with physical processes that control chloride deposition by correlation and regression analyses, and based on this to construct BCD map.

Line 16-17. Hypotheses (2) is mistaken. They are many references.
Line 17-19. This assumption (and relationship) was found and developed by several authors from the ninety (Rosenthal, 1988; Gasparini et al., 1989; Herrera and Custodio, 2008; Contreras et al., 2008; : : :) in other hilly coastal areas. This is not a novelty. Line 20-23. See comment on the Cl/Br ratio.
C2571

Discussion and actions
We don't think it matters whether a hypothesis is correct or not. It improves our understanding to test the hypothesis.We test the hypothesis with the data. This hypothesis is based on our understanding that wet deposition should increase with elevation due to increased precipitation. The correlation result weakly support this. For the terrain slope effect, thank for providing the references. But unfortunately, we don't find relevant

information related to our assumptions on topographic effect on atmospheric chloride deposition.

In Rosenthal 1987 JoH paper, coastal distance and bedrock type effects are discussed. Rosenthal 1988 JoH paper, no atmospheric chloride deposition is discussed. In Herrera and Custodio 2008, no relationship between groundwater salinity and elevation is found. Similarly in Minor 2007, chloride enrichment in spring is examined with elevation. It appears that two issues are mixed here. One is the possible topographic effect on atmospheric chloride deposition. The other is the link between chloride concentration in groundwater at different topographic locations. Our paper is addressing the first issue. In Contreras 2008, BCD measurements and elevation data are presented. The data suggest that a negative correlation between BCD and elevation. However, because coastal distance increases is highly correlated with elevation, it is hard to tell how much contribution from elevation to the BCD in their area.

Minor 2007 and Contreras 2008 works are now cited, which is relevant to the subsequent application of chloride mapping results or to our testing hypothesis.

Page 5856
Line 10-12. This detailed study requires to asses separately wet and dry deposition into bulk deposition before interpolate and derivate covariates. Line 10-13. Clarify wind stations in Figure 1.

Discussion and actions
Our data do not allow us to assess wet and dry deposition separately, although some partial correlation results infer something about the difference in wet and dry deposition in the area. Also, as we pointed out earlier, BCD (not the separate wet and dry deposition) is needed for CMB application. The wind sites are now described in the Figure 1 caption.

Line 15-20. You can asses land uses changes impact on groundwater resources for only 2-years long record of chloride deposition?. They are long series in the region?. Then, what is the expected natural variability of chloride deposition?.

Discussion and actions
Apparently, our writing in the abstract/introduction may have given you wrong impression that we are assessing groundwater recharge. No. What we discuss in this manuscript is BCD only. We agree that there is some inter-annual variability in BCD. But in comparison to spatial variability, inter-annual variability at each site is much smaller. This is why we include all data available in the area for the mapping purpose. Similar length or even shorter duration of samples is used in other studies (such as Alcala JoH 2008 v.359 189-207).

Line 23-25. Describe the analytical method to determine chloride in Laboratory, as well as accuracy and reproducibility. Line 28. Website of BOM.

Discussion and actions
We agree. The information is added.

Page 5857
Line 18. MATLAB software requires copyright?.

Discussion and actions
We have MATLAB license, and perform the analysis with MATLAB. We rephrase it.

Line 19. t-distribution. Describe better with references, examples and potential ranges. Line 21. r is rxy(z). Follow notation.

Discussion and actions
We agree. It is fixed. Reference is Lowry, 1999-2009.

Line 23. For t (Eq. 3), what units (supposedly dimensionless) and potential ranges are theoretically expected for uncorrelated and well correlated variables. All variables are linearly correlated. They are exponential and other type of correlation?. What is the weight of N (length of the series) in t?. Explain the relevance of t values and rxy(z) for short series with different N.

Discussion and actions
The value t is of no unit. This is standard student's t testing. Theoretically, there are infinite numbers of nonlinear correlation. Linear correlation is the first to be tested. N is the number of samples. The significance of t value is dependent of N. As Goovaerts pointed out, p-values from the t testing cannot strictly be used do the significance test because some spatial dependence in the data. This part is now rephrased, saying to use the p-values as illustration to find out which testing variables are more important than others for BCD, rather than strict statistical indicator.

Page 5858
Line 1-3. What may be the canopy effect on deposition (dry and wet separately). Natural or induced forest and stubble fires are nowadays frequents?.

Discussion and actions
This indeed is something we are looking at separately. Our preliminary sampling results suggest that chloride deposition under the canopy is significantly higher than the open area. As our samples included in this paper were all collected in the open area, canopy effect cannot be accessed. But we agree this is important issue to be examined. Not sure what bush fire effect would be on BCD. If we assume vegetation does not intake chloride, forest fire should not produce additional atmospheric chloride. Anyway, this is beyond the scope of this study.

Line 5-10. The procedure is probably satisfactory but its novelty limited. You can explain clearly the improvement of this method over the kriging with external drift, the co-kriging, etc. Kriging provide a suitable spatial error estimation with a clear meaning not found here.

Discussion and actions
The emphasis of this paper is not to justify the novelty of the method. Actually, this method has been published in 2005, in which, comparison between ASOADeK and co-kriging, as well as with PRISM, was performed. Please check our response to your general comment on the novelty of our paper. For the BCD mapping, several earlier studies were all relied on kriging, rather than CK or the KED. This is probably because it is difficult to find appropriate secondary variables to estimate BCD. The regression we defined with only two predictor variables interpret about 70% spatial variability of the 15 sampling sites. The contribution of this paper is for first time to apply ASOADeK to map BCD, and find the appropriate ASOADeK regression based on partial correlation analyses.

Line 6-7. Residual maps are a measure of spatial uncertainty?. Can you explain better this sentence?.

Discussion and actions

Residual here is the difference between the observations and the regression estimates. These residuals are regarded as a random variable, which is to be used for a kriging residual map. The residual map is then added to the regression estimate to obtain the final BCD map. This sentence is rewritten.

Line 12-15. Dry deposition measures are needed, as well as to study the source of chloride to identify predictable marine sources from punctual and/or regional anthropogenic or lithological sources largely dependent of wind intensity and periods of production (industrial factories) or urban pollution (winter and summer with probably most population by tourism activities and pollution admitted by see-breezes).

Discussion and actions

Dry deposition is something we desire to have. However, it is not available, and we don't think it is necessary for this study. If the relationship of dry deposition with a certain variable is different from wet deposition, the correlation between BCD and this variable is weak, and will not be included in the regression, such as the elevation factor that we examined. In terms of regression for ASOADeK mapping, separation of them is not necessary. In our study area, no obvious air pollution sources exist. The intensive anthropogenic activities locates in the coastal area in the west of the study area. If they release additional chloride source (unlikely) would not disturb the relationship we investigated in the correlation analyses. Actually, because of the irregular shape of the coastal line, some local marine aerosol source (such as that in the southwest of the study area) may affect the overall BCD relationship with the selected variables. This is why we exclude the two sampling sites in the correlation and regression analyses. We add some of this discussion in the text. Please also check our response to your general comments.

Line 24-25. Explain better sentences and definitions with details. What means b0. How is introduced cos (_-!) into Eq (4)?.

Discussion and actions

Line 24-25 is rephrased. Physically, b0 would be a lumped term, indicating the portion of total deposition that is uniform throughout the study area. One example can be the slow falling component in Keywood 1997 paper. Other terms are interpreted following Eq. (4). For details please refer to Guan 2005 paper.

Line 26. Only in southern areas?, not in urban or industrial areas?.

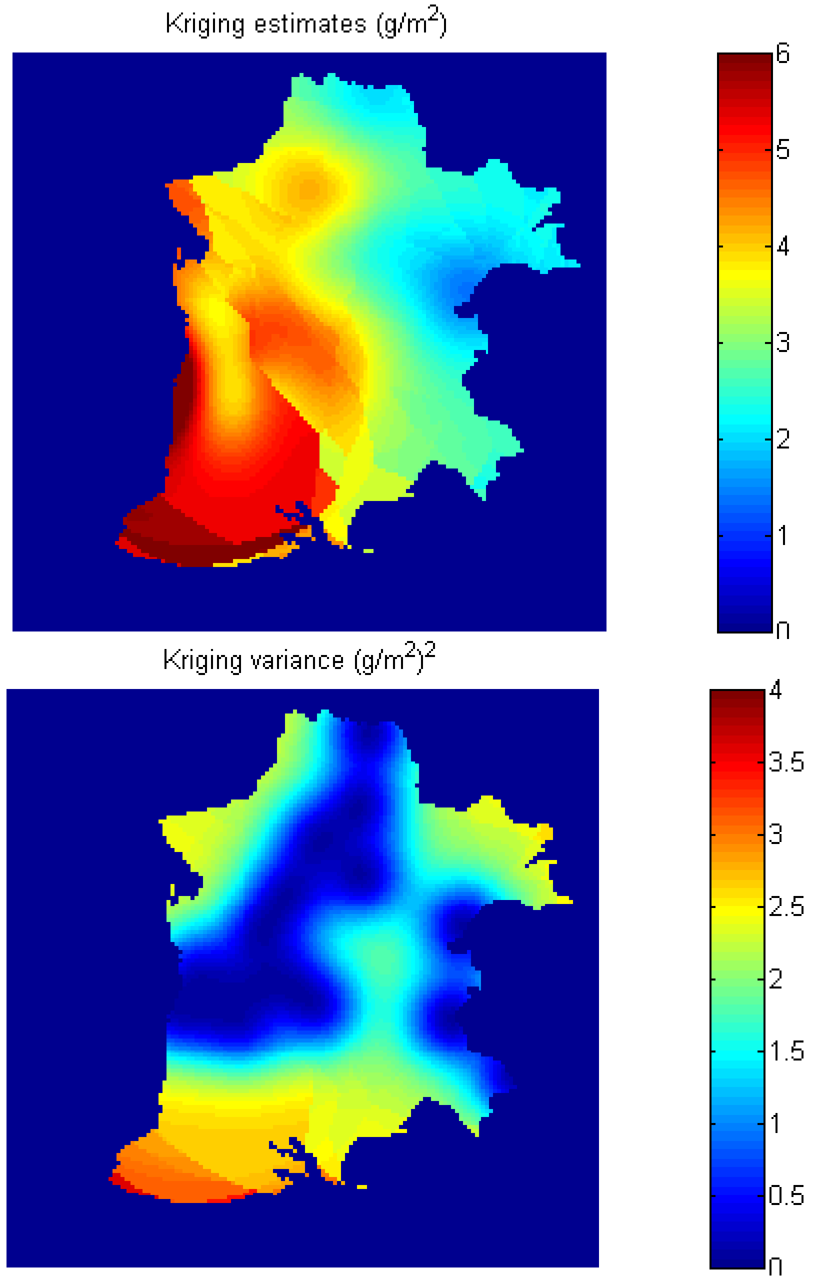
Discussion and actions

This is based on wind direction histogram. No evidence shows that urban area release additional chloride to the atmosphere in our study area. Please also refer to our earlier response to you similar comments.

Locate and rename stations in Figure 1 and Figure 3. Define density of BCD sites, as well as a simple BCD kriging variance map.

Discussion and actions

Figure 1 and 3 are improved. The direct kriging variance map is attached. We tend not to include it in the manuscript, because direct kriging is not our method. We would leave this for Editor to make decision.



Page 5859
Line 5-15. Justify divergences from bibliographic sources before exclude samples which are then necessary to interpolate and quantify spatial uncertainty.

Discussion and actions
We address this issue in our response to your general comments.

If dry deposition is too irregular, linear correlation methods should be excluded as suitable tool for regionalizing BCD. Probably, heterogeneity can be reduced grouping samples seasonally See Gustafsson and Larsson (2000).

Discussion and actions
With our data, it is difficult to assess dry deposition separately. As discussed earlier, this is not our focus for this paper. We could use seasonal BCD for the mapping if the atmospheric variables that we examine change in the year. But the prevailing wind for the precipitation is westerly throughout the year. Thus we decided to look at the annual BCD.

The ASOADeK method do not seems to improve estimation of BCD spatial variability in coastal areas.

Discussion and actions
The different performance of ASOADeK and kriging, over the same data, is summarized in figure 5. The cross validation results indicate the MAE is about 0.85 g/m^2, which is about 20% of average observed BCD in the area. The estimate error at 90% confidence level (1.645* standard error) is between 1-2 g/m^2, in comparison to kriging standard error (this is one, not 1.645) between 1-2 g/m^2 in the mapping exercise in Alcala et al. 2008 paper. And our mapping resolution is 1km *1km pixel, while it is 10 km * 10 km. in Alcala et al. 2008 paper.

Page 5860
Line 4-8. This is a well-documented scale effect. Probably you are sampling the first part of a regional exponential trend inland-ward. Line 9. Over that other coastal study areas?. See pioneer studies of Eriksson and Khunakasem (1969) conducted in Israel to the current studies based on GIS support (Minor et al., 2007).

Discussion and actions
We agree that we are sampling the part of exponential curve. The exponential decay of BCD over a large scale has been noticed for over decades in Australia (Blackburn and McLeod, 1983, Keywood et al., 1997), and we mentioned this in the text.

Line 10-13. You can include dry chloride deposition as independent term in equations to improve correlation, externally.

Discussion and actions
It works only when dry deposition data is available.

Line 21. They are references on canopy effect on bulk chloride deposition?. Line 23. This is evident and need to be assessed for mapping chloride deposition at that catchment scale.

Discussion and actions
We completely agree with you that canopy effect can be important. As our samples included in this paper were all collected in the open area, canopy effect cannot be accessed. Please refer to our response to your earlier similar comments.

Page 5861
Line 15. Where is the distance between first rainwater samplers and the coastline?.
Line 17. Gradients of 2 g m-2 year-1 km-1 reported by Alcalá and Custodio (2008) refer only for spurious data adjacent to the coastline in northwestern Iberian Peninsula. The rest of the data are between 0.05 and 0.5 g m-2 year-1 km-1, and usually between 0.05 and 0.25 g m-2 year-1 km-1 in the Mediterranean non polluted coastal areas (see Figure 7 in Alcalá and Custodio 2008).

Discussion and actions
The first sampling site is about 3 km from the coast. Thanks for your reminder of the detailed work in Spain. Then, the result from our regression is within the ranges of Alcala and Custodio' study. This is rephrased.

Line 27. Show range of MAE relative to D measures. It is low for coastal places but very high for inland stations (up to 50%). Explain.

Discussion and actions
The estimate MAE data is shown in Figure 5. The AE is shown in the scatter plot. MAE is the average of all data point AE. Our method does not show different estimate error at the lower BCD sites. The uncertainty map is derived from kriging variance and regression standard error maps. The high mapping uncertainty is resulted from the sparse sampling sites in the inland area.

Page 5862
Line 11-13. Define equation for mapping uncertainty with examples and references

Discussion and actions
We agree. The text and equation shown below are added.

The mapping uncertainty (ε) is composed of the regression uncertainty and kriging uncertainty. With an assumption that the mapping uncertainty follows normal distribution, it is calculated as

$$\varepsilon = u\sqrt{\varepsilon_r^{\ 2} + V_k} \qquad (5)$$

where *u* is the critical value of the standard normal distribution, (1.645 for 90%, and 1.960 for 95%), ε_r is the standard error of the regression fit, and V_k is kriging variance.

Line 20. Conservative?. Explain better. May be a mistake in error propagation equation?. Define error equation. Line 29. Explain better the sentence.

Discussion and actions
We improve the uncertainty mapping. See our response to your previous comment.

Page 5863
Line 5-6. Scanlon (2000) studies the Eagle sedimentary basin. This is an inland flat area. The relations are reliable?.

Discussion and actions
We believe so. With the assumption that the atmospheric input of chlorine-36 is uniform over the whole area (smaller in terms of spatial variability of chlrorine-36 deposition), the atmospheric chloride deposition can then be estimated from the ratio of chloride over chlorine-3. The 30% uncertainty is from the uncertainty in estimating chlorine-36 fallout. Actually, Scanlon recommended us to sample chlorine-36 data in our study area, but we have not started the sampling plan yet.

Line 15. It is a well-know rule in most coastal zones.
It is not compensated by elevation but the canopy effect. Several papers focus the atmospherically contributed nutrients to the land through wet and dry depositions. Line

Discussion and actions
Do you mean wet deposition is enhanced by elevation? From our preliminary sampling (not included in this study), canopy may enhance deposition, but, is highly variable in space. As our samples included in this study were collected in the open area, canopy effect is not embedded in the data. We make this point clear in the study.

25. This assumption was demonstrated in many papers. At the rainfall event starts chloride increases by washing the initial chloride-rich see breezes and then chloride decreases: : : They are information on sea-waves size in addition to the wind intensity?.

Discussion and actions
Thanks for the reminder. We had not seen published data. Our own data (Figure 8) support this assumption. Apparently, wind is an important player in generating marine aerosol and transporting chloride into inland. It would be good to have high temporal resolution data to reveal this.

Page 5864
Line 10-28. This is a speculative sentence with light implications, without new contents of general interest or data acquisition, formulations and CMB method applications or improvements. Discussion should be improved with the contribution of the paper.

Discussion and actions
The slope orientation (terrain aspect) effect is one significant point inferred from the regression results. The discussion on line 10-28 is to provide a likely physical mechanism, based on data. But we agree, our analysis may not reflect the actual physical processes. We add another optional mechanism to interpret the inferred terrain aspect effect, and simplify the previous discussion. The optional one is that recycle dust at the dry eastern flank may artificially increase the atmospheric deposition.

Page 5865
Line 1-15. Out the scope to calculate groundwater recharge through the CMB method

Discussion and actions
Again, this is to find out the potential mechanism that the terrain aspect becomes a significant factor influencing BCD distribution. We make the whole paragraph concise.

Line 8-9. If they are air pollutants in the area, they can be evaluated to justify low correlations in variables.

Discussion and actions
We cannot agree with you more on this point. We follow the same idea to develop our regression, and based on which we removed site #16 and 17 from the correlation and regression analyses.

Figure 1 need a large improvement. 4 maps are needed (1) regional location in Australia;
(2) a good quality contour map with places cited in the text, rivers, coastline, etc.;
(3) precipitation map; (4) 1-km DEM; (5) aquifer contour and a representative section.
Geographical coordinates, etc: : : Clarify wind and BCD stations.

Figure 7. The Peninsula of Gulf of St Vicent is cut in b through f and data are missing.

Discussion and actions

Figure 1 is now significantly improved, following most of your suggestion. The aquifer contour is not necessary for this paper, thus we don't add it. The missing part in Figure 7 b through f is because residual kriging does not provide estimates for that area due to lack of sampling sites.