

Dear Dr. Crosbie,

Your comments and suggestion, especially your calculation on coastal dependence function, are greatly appreciated. Below are our responses. The marked manuscript revision, on top of Nov-24 revised copy for the previous three reviewers, is attached. Look forward to your further suggestion.

Sincerely,

Huade Guan
On behalf of the co-authors.

General comments

(1) This manuscript fits a statistical model to chloride deposition data in the Mount Lofty Ranges to enable a spatial surface of chloride deposition to be produced. A spatial surface of chloride would be very useful as an input to estimating groundwater recharge on a regional basis using the chloride mass balance. The manuscript aims to test the hypothesis that elevation, slope and aspect are important factors in chloride deposition.

Discussion

Yes, our aim is to find out the factors influencing chloride deposition in the MLR, and based on this information to construct a chloride deposition map. Besides the well-known the distance to the coast (referred to as coastal distance in the text), topographic factors (elevation, slope and aspect) are examined.

(2) The major problem with this paper is that it assumes that easting is an appropriate covariate for predicting chloride deposition. The reasoning given for this assumption is that the wind comes from the west so therefore the chloride should too. To make this model fit the data, two data points (Murray Bridge and Tailem Bend) were excluded because they did not fit the trend with another data point (Mannum). The reason they did not fit the trend is because they are closer to the Southern ocean than St Vincent Gulf (see Figure i below).

If distance to the coast is used to predict the chloride deposition (for all 17 points) rather than the easting the model fit is much greater (see Figure ii below). The r^2 for distance from the coast is 0.711 compared to 0.620 for the easting. In previous studies of chloride deposition in Australia (e.g. Keywood et al) an exponential or double exponential decay function with distance from the coast has been used. If this model is used with the data set presented in this manuscript then the r^2 is 0.759. This simple model is a better fit to the data than the ASOADek model presented.

I cannot recommend this manuscript for publication as the methodology is flawed and the conclusions cannot be fully substantiated. Some detailed comments are below.

Discussion

We agree with you that the coastal distance is a primary factor controlling chloride deposition, revealed in Keywood et al. 1997. Blackburn and McLeod (1983) earlier observations also suggest similar exponential decay (although no explicit equation is given)

in atmospheric chloride deposition when one progresses inland. These observations support that (1) atmospheric chloride originates from the ocean, and (2) it is transported inland by moving air (wind) and cloud and falls out to the land surface by wet and dry deposition. However, it is difficult to directly apply coastal dependent relationship in chloride deposition mapping. There are two difficulties. First, due to coastal distance dependence is physically resulted from how efficiently the atmospheric chloride is moved inland, and how quickly it falls out or is precipitated out, the function parameterization is different from places to places (Alcala et al., 2008), because of the difference in wind and precipitation climate. Second, it is difficult to determine coastal distance because this distance should be calculated from the coastal point that is upwind from the mapping pixel. And it is computationally expensive (if not impossible) to determine coastal distance for each mapping pixel. This is probably why that this coastal distance relationship has not been used in detailed chloride deposition mapping that we reviewed in the introduction section.

In our paper, we attempt to incorporate this coastal distance dependence into the mapping, using the ASOAdEK model. The idea is that in a small area where the wind and precipitation climate is relatively simple, it is likely that one parameterization of coastal dependence function can be applied. Our earlier work (Guan et al. 2009 in J. of Hydrology, table 2, attached in the appendix) suggest that the moisture flux direction for precipitation in the study area is westerly to southwesterly. This should be similar to prevailing wind direction when chloride wet deposition occurs. For the dry deposition, we examine wind direction frequency, and find that westerly (although not exactly 270 degree) is the wind direction that most likely to transmit marine aerosols into the MLR (Figure 9), especially in winter season. Thus, it is likely we can lump the wet and dry deposition together in term of coastal distance dependence. But, as you mentioned, the problem is that both St Vincent Gulf and Southern Ocean could provide marine chloride aerosols for the study area. For the aerosols from St Vincent Gulf, as the coastal line is almost parallel to the longitude, the UTM easting X is linearly related to coastal distance, and can be used as a proxy of coastal distance for linear correlation and regression analyses. But for the Southern Oceans source, we agree with you that X is not an approximate quantity for coastal distance. For the relative position between the study area and Southern Ocean, the UTM northing Y can be good approximation for coastal distance with marine aerosols for this source. Another problem is that it is very unlikely to use one coastal distance dependence function to fit chloride deposition from two sources (we will discuss this issue with your linear correlation analysis later). Fortunately, the chloride source from Southern Ocean does not affect the most part of the study area (this is supported by low partial correlation coefficient between D and Y). From wet deposition view point, westerly or southwesterly prevailing wind direction, with source from Southern Ocean only affect the southeastern corner of the study area. In terms of dry deposition, the westerly wind in the study area corresponding to transporting St Vincent source chloride into the area is more dominant than the southerly wind corresponding to Southern Ocean source. Thus, we believe it is reasonable to focus on the chloride deposition from the primary source (from the west). Based on this, the two sites in the southeastern corner of the study area are excluded from regression analysis which is used to produce the overall chloride deposition trend for the whole study area, but they are included in the residual kriging which to mainly collect the southern ocean source chloride (or other local source chloride) into the deposition map.

Your linear regression analysis between chloride deposition and the coastal distance gives high R square. It seems to tell that one coastal distance dependent function can be used to describe chloride deposition from the two sources in the whole area. But careful examination tells that coastal distance from the Southern Ocean is calculated only with two data points, in comparison to 15 points for which the coastal distance is measured from the St Vincent Gulf coastline. Even if it is luckily that chloride deposition from the two sources can be described using one coastal dependent function, due to the difficulty to incorporate coastal distance directly into the mapping procedure as discussed above, we cannot apply it directly in the

mapping. Thus, we would rather focus on the primary chloride source to the area, and treat the two sites (#16 and 17) separately. For this small area, linear relationship and exponential relationship, between chloride deposition and coastal distance, is similar. We decided to use the simple linear relationship.

The final ASODeK model estimated chloride deposition has an R square of 0.85 (please check Figure 6b in the revised manuscript, or Figure 5b in the original manuscript) with the observation, although with increased complexity.

Actions

Apparently, we have not written the text clearly enough. Some of the above discussion is now included in the revised text. Please check our marked revision.

Specific comments

(1) P5853, L14 CMB is most commonly used on the plains and rarely used in mountainous terrain. Runoff adds considerable complexity to the method. No justification is given as to why the CMB is appropriate in mountainous terrain.

Discussion and actions

We agree that one-dimension CMB method is more commonly used in the basin floor. When CMB method is applied in mountain terrain, stream output of chloride should be included in the CMB equation, as described in equation (1). Here we mean that CMB provides a good solution to estimate groundwater recharge in the mountain areas. We don't intend to state that CMB is more commonly applied in mountains than basin floors. The sentence is slightly rephrased.

(2) P5854, L12 The siting of samplers in the open will underestimate chloride deposition because of impingement and entrainment of dryfall in the vegetation. How is this incorporated?

Discussion and actions

We totally agree with you about the potential canopy effect on chloride deposition. We are actually conducting field experiments to examine the canopy effect under the native and planted vegetation. However, due to the nature of the available sampling data for this manuscript, we cannot access canopy effect. This issue is made clear in the revised manuscript.

(3) P5855, L13-8 How were the sites selected? There is no detail given on the elevation, aspect and slope of the sites even though the hypotheses to be tested are that elevation, aspect and slope are determinants in chloride deposition. The experimental design does not appear to be adequate to answer the questions posed. There are no mentions of transects up a constant slope or sites with equal elevation but different aspects.

Discussion and actions

Our study is based on the existing chloride deposition data. Part of the data was collected by co-author Kayaalp at Flinders University, along a transect from the coast, across the Ranges, to Murray River. The other is collected by DWLBC primarily to estimate chloride deposition in the mountains. Ideally, the data collection should be designed and performed following the question and hypothesis. For this study, we use existing data to test our hypothesis. Of the 15 sites, five sites have an eastern aspect, and 10 sites have a western aspect. The site elevations spread from below 50 meters to above 500 meters. The coastal

distance spreads from a few km from the coast to the eastern edge of the study area. We believe this is a fairly good data set for our purpose.

The idea of setting up site pairs to examine the hypothesis is difficult to apply in this case because besides topography, coastal distance strongly influence chloride deposition. Thus, following your idea, we should select two sites with exactly the same coastal distance and elevation, but on two opposite slopes. This is difficult. Even if we find some points with similar coastal distance and elevation, it is still difficult to choose the sites, because we don't know in advance on what spatial scale the terrain aspect is effective. For precipitation, we find that terrain aspect in 13 km scale influences precipitation (Appendix I), while for chloride deposition, the result infers that the effective spatial scale is 7 km.

Some of the above information is included in the revised Table 1.

(3) P5855, L13-8 Why not include precipitation? Is it correlated with elevation, slope and aspect?

Discussion and actions

We examined relationship between chloride deposition and precipitation in correlation analysis, but didn't list it as a hypothesis to test. This is because precipitation is correlated with elevation, slope and aspect (Appendix I of this document) too.

(4) P5856, L10-4 Fig 2 does not support the assertion that both wet and dry deposition occur from the west. In Fig 2, only 2 of 8 plots show that the wind from the west is greater than the other directions. This is a key assumption in the methodology and it cannot be substantiated with the data shown.

Discussion and actions

For the wet deposition, the westerly direction is inferred from the analyses of 96 gauge long-term precipitation observations (Appendix I). For the dry deposition, we only compare marine aerosols from the two sources, westerly from the St. Vincent Gulf, and southerly from the Southern Ocean. If the dominant wind from the north, it would not contribute to transmit marine aerosol inland, such as the case at site 4 of figure 2. Thus, in terms of transmitting marine aerosol to the study area, we only need to compare westerly and southerly wind, and see which one is more frequent. Of the four sites, westerly dominant wind is observed for two sites, while none is observed with dominant southerly wind. To support our statement, Fig 9 is cited in the revised manuscript. The text is also slightly rephrased.

(5) P5857, L6-8 An r value close to ± 1 does not indicate a physical causal relationship between the variables, it only implies there could be one. The language used in this sentence is far too definite.

Discussion and actions

This sentence is removed.

(6) P5859, L22 It is an assumption that the chloride comes from the west, not a fact.

Discussion and actions

This statement is slightly rephrased.

(7) P5859, L22-3 The "abnormally high" results come from the most southerly points. These are not outliers to be discarded because they do not fit the model. They highlight the deficiencies in the model. These points are closer to the Southern Ocean

that St Vincent Gulf, could it be that this is the source of some of the salt? The assumption that all salt comes from the west may not be valid for these points.

Discussion and actions

What you said was more or less included in our text. Probably the word “abnormally high” is not appropriate here. It is rephrased.

(8) P5860, L3 Keyword et al used a double exponential function for the relationship between chloride deposition and the coast to allow a much more rapid decrease in deposition close to the coast. Why was this model dismissed without trialling it?

Discussion and actions

We did test the exponential function of X. It gives similar result. Over a short distance, a linear decay in chloride deposition rate is a good approximation for the exponential decay (Alcala et al., 2008). This issue is mentioned in the text.

(9) P5860, L8 How can this be a “fact” if no alternatives were considered? Is this the best model or merely the one used for the study?

Discussion and actions

We agree. It is not appropriate to use “the fact” here. The word is removed in the revised manuscript.

(10) P5860, L22-7 How can dry deposition be extracted from bulk rainfall samples when it was not sampled for in the first place? I am not convinced that this is a valid analysis. The effect of precipitation upon deposition is removed and then the residuals are correlated with the distance from the west and this is proof that dry deposition is dependant upon distance from the west. There is no correlation between precipitation and deposition (tab 2), this has been shown many times because increased precipitation leads to a lower concentration of chloride in precipitation resulting in little change in deposition. By removing the effect of precipitation it is only noise that is removed and the original relationship remains – chloride deposition is correlated with distance to the west.

Discussion and action

We agree with you on this point. By removing precipitation effect in the partial correlation analysis is to exclude the precipitation amount effect on chloride deposition. Our earlier thought to use it to separate the effect on wet and dry deposition is not correct. This part is rewritten. Your insight into this issue is greatly appreciated.

(11) P5863, L11 – P5864, L9 Wet and dry deposition cannot be separated using this argument. On an inter-event basis there is often seen an inverse relationship between chloride concentration in rainfall and rainfall amount. This indicates that the atmosphere only holds a certain amount of chloride that can be rained out, big rainfall events are dilute, small ones are concentrated. This intra-event comparison has nothing to do with elevation.

Discussion and actions

This part of discussion is different. Here we discussed why D and Z have lower partial correlation coefficient. We didn't to separate wet and dry deposition, but to discuss D and Z

association from wet and dry deposition aspects separately. Anyway, the part is rewritten slightly.

(12) P5864, L15-6 “we find that due to land-sea wind circulation, westerly and easterly winds frequently occur within a day” The author has provided proof that their assumption that chloride comes from the west does not always hold true.

Discussion and action

Apparently, you assume that the wind, no matter where it is from, always carries marine aerosols. Otherwise, you won't come to this conclusion. Please refer to our response to your comment (4).

(13) P5864, L21-2 How can 9.00 am and 3.00 pm represent night-time and daytime when neither of them is during the night? This whole paragraph is pure speculation and actually diminishes confidence in the findings of the work. It should be deleted.

Discussion and action

Although the measurements are conducted at two time points, with the understanding the physics of land-sea circulation, in coastal area, day-time wind can be roughly represented by 3:00 pm observations, while night-time wind can be roughly estimated from early morning observation. In most of the day time, sea breeze occurs due to faster warming up at the land surface in comparison to the sea. The temperature difference persists throughout the day (usually 11:00am through 5:00pm, Thomas Foken, Micrometeorology, p226). Similarly, seaward land breezes develop soon after sunset, although much weaker and slower than sea breeze.

Anyway, this part of discussion is made much more concise in the revised manuscript. And the word “night time” is removed.

(14) P5865, L14 Precipitation in summer is comparatively small over the study region. Perhaps an analysis of the deposition of chloride in summer and winter would shed some light on the wet vs dry fall argument?

Discussion and action

This is interesting, and should be explored in the future. If we assume that at one location, chloride concentration in the event rain water is similar in winter and summer, analysis of separate seasonal bulk chloride deposition and precipitation, can be very helpful. But this is beyond the scope of this manuscript. L14 is removed.

Appendix I: ASOADeK inferred moisture flux direction (MFD) for the MLR, from Guan et al., 2009 in J. of Hydrology, which is cited in the manuscript.

Table 2
The results of ASOADeK regression with 12 monthly precipitation in the study area (symbols are listed in Eq. (2)).

Month	Window ^a (km)	b ₀ (mm)	b ₁ (mm km ⁻¹)	b ₂ (mm km ⁻¹)	b ₃ (mm km ⁻¹)	b ₄ (mm)	b ₅ (mm)	Adjusted ^b R ²	MAE ^c /mean (%)	MFD ^d	MG ^e
1	13	21.1	-0.007	-0.051	27.01	-43.54	-52.05	0.80	6.9	230	187
2	15	21.4	0.012	-0.062	24.63	-30.29	-58.75	0.53	9.6	243	169
3	15	25.9	0.019	-0.132	41.06	-8.67	-119.18	0.79	9.3	266	172
4	11	53.3	-0.108	-0.207	86.66	-94.79	-154.68	0.91	7.8	239	208
5	13	77.0	-0.136	-0.346	143.27	-84.68	-326.74	0.89	8.9	255	201
6	15	94.3	-0.200	-0.438	182.54	64.37	-350.94	0.89	9.7	280	205
7	13	98.7	-0.287	-0.425	195.60	-130.46	-274.82	0.91	9.6	245	214
8	15	77.9	-0.116	-0.362	182.25	-20.17	-299.85	0.90	8.9	266	198
9	13	62.3	-0.156	-0.171	137.52	-230.05	-200.74	0.90	8.1	221	222
10	13	49.2	-0.084	-0.129	93.65	-156.61	-168.37	0.89	7.6	227	213
11	13	29.7	0.004	-0.099	51.56	-114.77	-141.26	0.83	7.9	231	178
12	13	25.0	0.026	-0.088	39.33	-58.06	-96.29	0.82	7.8	239	164

^a The optimal DEM window size for calculating terrain aspect and slopes for the regression.
^b Adjusted coefficient of multiple determination considering the number of predictor variables effect.
^c MAE is the regression mean absolute error.
^d Prevailing moisture flux direction, zero from the north, increasing clockwise, 180 from the south.
^e Atmospheric moisture gradient, zero indicates wetter in the north, increasing clockwise, and 180 indicates wetter in the south.