Response to Reviewer Comments – Reviewer 1

We thank the editor and the three reviewers for their constructive comments and suggestions. We thank all three reviewers' comments that the text is well written and for their recognition of our study as a valuable resource for groundwater practitioners. We believe that in addressing their comments, the manuscript will be considerably improved and be ready for publication.

Most questions were about minor text updates and queries. Two reviewers asked for further comparisons between our outputs and previous investigations. We present a suggested approach to address these comments, including new figures for both the manuscript and the supporting information.

We believe that these additions directly address reviewer concerns, clearly showing the impact of the distribution of our estimates as a primary control on the differences in recharge estimates between our study and previous studies.

Our responses to the reviewer's comments (**RC**) are provided below as author's comments (**AC**). To help with the assessment of our responses, we colour coded our responses into agreement (green), partial agreement (yellow) and disagreement (red). When referring to text excerpts in our manuscript, we have provided the line number and whether text has been removed, or if new text is added.

RC1: This paper presents an interested method to estimate groundwater recharge rates across Australia using chloride measurements. The text is well written and logically organized so that is easy to follow. The figures are excellent and ready for publication. I am not specialist of geochemistry or chloride but I found this study very relevant for hydrologist like me who is interested in groundwater processes. While I consider my following comments as minors, I think they must be addressed with attention before publication.

AC: Thanks for your interest, positive feedback and helpful comments on our manuscript that intend to improve our work.

RC2: I am especially disappointed by the comparison between the presented product (from chloride) to the previous estimates from other studies (for example Moeck et al. 2020). Such previous studies report mean recharge estimates 5 times larger than the present study. You only attribute this drastic difference to "the spatial distribution of recharge point estimates, and the estimation of recharge values using different recharge estimation techniques" (line 470). I am sorry but I am not very convinced by this argument.

AC: We partially agree (minor changes to the manuscript and supporting information suggested). To address this point, we will provide updates to the text (see below) and present a new figure in the manuscript and another in the supporting information to make this point more definitively. See suggested text changes, figure and supporting information figure below.

Suggested text and figure changes in manuscript

Suggested revision at line 469: The higher mean recharge values of the point data reported in other studies that cover Australia (e.g., Crosbie et al., 2010a; Moeck et al., 2020; Berghuijs et al., 2022) compared to ours can be attributed to the difference in spatial distribution of recharge point estimates, and the different recharge estimation methods used. estimation of recharge values using different recharge

estimation techniques (i.e., water table fluctuation method, water balance, CMB, and other tracers). Several differences in the method are important, including:

(1) 60 % of the estimates in Crosbie et al. (2010) and Moeck et al. (2020) were from an earlier study (Crosbie et al., 2009), which used a simpler CMB method and an older chloride deposition map to calculate recharge (see chloride deposition maps suggested for the supporting information, figure S9b).

(2) Our method incorporates the most recent improved chloride deposition map with enhanced data and spatial coverage (Wilkins et al., 2022).

(3) There are key differences in chloride deposition rates between the different chloride deposition maps, especially within 50 kilometres of the coastline, that can significantly affect the resulting recharge rate (see chloride deposition maps suggested for the supporting information, figure S9).

The mean of the 2,722 CMB recharge estimates from Crosbie et al. (2009) is 388 mm y⁻¹ while the mean of the other 1,620 estimates from Crosbie et al. (2010) which were estimated from 14 different methods (including 38 % from CMB, 25 % from transient soil CMB, and 9 % from water table fluctuation) is 40 mm y⁻¹. The estimates from Crosbie et al. (2009) are likely overestimates and were flagged by Crosbie et al. (2010) to have very little quality control.

(4) Our approach accounts for chloride lost to runoff in the calculation, resulting in a reduction in our recharge rates compared to the simpler method used in Crosbie et al. (2009) which does not consider this factor.

(5) Our methodology is stochastic, performing 1,000 recharge calculations to generate a probability distribution. We present the median and an error range taken as the 5th and 95th percentiles of the distribution to provide a more robust interpretation of the results.

The spatial distribution is important because the climate at the location of the recharge estimate strongly influences the annual recharge rate (Moeck et al., 2020). Figure 7 (new, below) demonstrates this point by using the climate zones found in Australia that are classified from different aridity index values (i.e., in order of increasing aridity or decreasing recharge potential: humid, dry subhumid, semi-arid, arid, and hyper-arid, based on United Nations Environment Programme, 1997).

Suggested new figure (Figure 7) is shown below:



Figure 7 Maps and histograms showing the difference in spatial distribution and proportion (%) of the point recharge dataset of (a, d) Crosbie et al. (2010a), (b, e) Moeck et al. (2020) and (c, f) our study that are located in various aridity classes (Hyper-arid, arid, semi-arid, dry-subhumid and humid; United Nations Environment Programme, 1997). The proportion (%) and mean recharge (mm y^{-1}) are shown in the histograms above each bar.

The proportion of recharge estimates from Crosbie et al. (2010a) and Moeck et al. (2020) located in dry subhumid and humid aridity classes is double that of our dataset (Figure 7). The mean recharge rates in Crosbie et al. (2010a) and Moeck et al. (2020) for each aridity category are all higher than our study – particularly dry subhumid and humid which are 3-4 times higher. The higher proportion of estimates in the dry subhumid and humid climate zones together with the significantly higher mean recharge rates in these climates, results in a higher overall mean recharge rate for the Crosbie et al. (2010a) and Moeck et al. (2020) datasets compared to our study.

Suggested revision at line 492: Our gridded map contains 278,253 pixels of which ~80 % are in an arid Köppen-Geiger climate (see Figure S11 in the supporting information), compared to ~26 % of the global land area that is classified as arid (Gaur and Squires, 2018).

Suggested figure in supporting information

Suggested new figure S9 (supporting information) shown below:



Figure S9. Maps showing gridded deposition maps from (a) Davies and Crosbie, (b) Crosbie et al. (2009), (c) Wilkins et al. (2022) and (d) Wilkins et al. (2022) and recharge sites that were able to be matched (green) and unable to be matched (red) to those in our study.

RC3: Why you do not compare point estimates from Moeck et al. (2020) with your product at the same locations, exactly? For example, using a squatter plot, or the density function of each product. In other words, I find your comparison with existing previous products is not enough detailed. Please, improve this comparison.

AC: We partially agree (minor changes to the manuscript and supporting

information suggested). Only a limited comparison can be made for several reasons, outlined below. This discussion will be added to the supporting information, with associated analyses.

Suggested text changes in manuscript

Suggested revision at line 472: Further details including limitations on the comparisons with Crosbie et al. (2010a) and Moeck et al. (2020) are provided in the supporting information. Studies that collated recharge estimates from other continents have also reported higher recharge rates than our point estimates.

Suggested text and figure changes in supporting information

Suggested new text and figure (S10) in the supporting information shown below:

6. Comparison of our point dataset with Moeck et al. (2020) and Crosbie et al. (2010a)

Only a limited comparison can be made between our point recharge dataset and Moeck et al. (2020) and Crosbie et al. (2010a) due to the following reasons:

1) The Moeck et al. (2020) dataset did not provide information on the estimation method or bore IDs, and only approximate location information. Thus, identifying specific bores to allow like-for-like analyses is not possible. Only 346 out of 4,579 Moeck et al. (2020) estimates could be approximately paired with a bore using matching latitudes and longitudes.

2) However, the Crosbie et al. (2010a) study (data contained within the Moeck et al. (2020) dataset) collated a dataset of Australian recharge estimates (n=4,360), presenting the data in a spreadsheet. This spreadsheet also included the recharge estimation method/technique used in the original study as well as in some cases, the bore ID.

3) Approximately 60 % of the Crosbie et al. (2010a) dataset was collated from Crosbie et al. (2009). These recharge estimates were flagged by Crosbie et al. (2010a) to have very little quality control. Upon review, we found that their methodology was too different for a meaningful comparison with our study (e.g., they utilise a simpler CMB method with a vastly different and recently revised chloride deposition map). I.e., we utilise recent chloride deposition maps from Wilkins et al. (2022).

For our comparison, we utilise the remaining data from Crosbie et al. (2010a), which comprise datasets from four recharge studies (i.e., Banks et al., 2007a, b; Green et al., 2007; Harrington et al., 1999). Figure S10 compares the recharge rates from these four studies to those matching in our study.



Figure S10. Comparison of recharge rates collated for Crosbie et al. (2010a) against median recharge rates (R_{50}) from our study. Grey open circles represent the mean recharge rates from Banks et al. (2007a) which plot closer to the 1:1 line than the minimum recharge rates collated in

Crosbie et al. (2010a). Blue arrows show the deviation of Green et al. (2007) recharge rates from the 1:1 line due to the different chloride deposition values used in their study.

The recharge estimates from (Harrington et al., 1999) and (Banks et al., 2007b) had similar recharge rates compared to our study (plotting close to the 1:1 line). However, our estimates were consistently higher compared to estimates from (Banks et al., 2007a). Our estimates were higher than those from the (Banks et al., 2007a) study, as Crosbie et al. (2010a) used the minimum estimates from that study (Fig. S10, solid grey circles. The mean recharge estimates (grey open circles, Fig. S10) effectively lie on our 1:1 line. Similarly, our estimates are consistently lower than those from Green et al. (2007). The large difference between our estimates and Green et al. (2007) can most likely be attributed to their methodology for calculating chloride deposition rates for their study. Further discussion below.

Green et al. (2007) used either an annual rainfall value of 800 mm y⁻¹ or 840 mm y⁻¹, along with a rainfall chloride concentration of 7.2 mg L⁻¹ or 8.4 mg L⁻¹, equating to a range of chloride deposition values between 57 kg ha⁻¹ y⁻¹ and 70.6 kg ha⁻¹ y⁻¹. The chloride deposition values used in Green et al. (2007) are approximately double those used in matching bores from our study, which range from 26.1 kg ha⁻¹ y⁻¹ to 38.0 kg ha⁻¹ y⁻¹. Their average rainfall chloride concentrations were calculated from 3-4 rainfall samples collected over a two-year period, making their chloride deposition rates less reliable. Our chloride deposition rates have been spatially extrapolated from rainfall gauges with a larger number of samples. Additionally, the runoff coefficients used in our study which ranged from 0.1 to 0.39 (average of 0.24) tended to be higher than the 0.1 blanket value used in Green et al. (2007). Both factors have most likely contributed to the recharge rates in Green et al. (2007) being double those calculated in our study.

RC4: It seems that, naturally, Australian soils are largely affected by salt (Wicke et al. 2011; https://www.encyclopedie-environnement.org/en/zoom/land-salinization/). Perhaps my understanding is not very good, but it seems that only chloride range of 35-125 mg/L for groundwater is considered as normal. Your Figure 1 shows that at least the half of your data are superior to this range. Could your underestimation (of recharge estimates) thus be due to a natural large groundwater concentration in chloride (Clgw) making your equation 2 obsolete?

AC: No change suggested. We are unsure where the reviewer has obtained the chloride range of 35-125 mg/L from as it is not in the link provided. Groundwater in Australia commonly has high Cl concentrations largely due to moderate rainfall, the semi-arid climate, and high transpiration rates of the native vegetation (Allison et al., 1990; Cartwright et al., 2004), which results in most of the water being returned to the atmosphere and little recharge (which is what our analysis shows). Our analyses use ~100,000 measured values from a database managed by an Australian government organisation. We provide a detailed description of the process to omit unreliable data values.

RC5: Chloride concentration in groundwater could be significantly impacted by human activities like agriculture or industry. If anthropogenic chloride "flows" into groundwater, its concentration will be larger than in natural systems and then, because equation 2, your estimates will be biased and too low. Are you sure that your measurements are not drastically impacted by these processes?

AC: We partially agree (minor changes to the manuscript suggested). The reviewer correctly highlights that additional sources of chloride may cause our estimates to be lower. While salting of roads is not common practice in Australia, anthropogenic sources of chloride such as from irrigation water and fertilisers may alter groundwater chloride concentrations. Additionally, the majority of studies where chloride/bromide (Cl/Br) ratios are reported indicate that the Cl is from evaporation of rainfall and not from other sources, which would lead to elevated Cl/Br ratios.

Suggested text changes in manuscript

Suggested revision at line 508: By not excluding bores located in alluvium, point and modelled recharge estimates for these bores can be underestimated if additional chloride not sourced directly from rainfall is present, for example, through the application of irrigation water or chloride-based fertilisers (e.g., potassium chloride).

RC6: Line 550, you clam "Our CMB-based recharge rates are considerably lower than other studies including global water balance models (e.g., Döll and Fiedler, 2008; de Graaf et al., 2015; Müller Schmied et al., 2021). This is likely due to the fact that global water balance models estimate modern recharge." ... Ah ok, but why, I do not understand your explanation? Do you mean that your estimates do not account for modern recharge, so they account for what? In other words, what is the period of validity of your estimates?

AC: We partially agree (minor changes to the manuscript suggested). We will address this comment in three ways: (1) We will include a new figure in the manuscript (Figure 7) to show that our dataset contains significantly more recharge estimates in the arid and semi-arid zones than other datasets, (2) we will include the text below relating to the timescales of CMB recharge estimates, and (3) the text below relating to land use change and the impacts on recharge estimation methods that operate on modern timescales.

Suggested figure and text changes in manuscript

(1) Suggested new figure (Figure 7) shown below:



Figure 7 Histograms and maps showing the difference in spatial distribution and proportion (%) of the point recharge dataset of (a, d) Crosbie et al. (2010a), (b, e) Moeck et al. (2020) and (c, f) our study that are located in various aridity classes (Hyper-arid, arid, semi-arid, dry-subhumid and humid; United Nations Environment Programme, 1997). The proportion (%) and mean recharge (mm y^{-1}) are shown in the histograms above each bar.

(2) Suggested revision at line 550: This is likely due to the fact that CMB operates on longer timescales that span the residence time of the groundwater (e.g., chloride can take between 4,000 and 40,000 years to accumulate in the Murray Basin, South Australia).

(3) Suggested revision at line 550: Contrary to this, global water balance models estimate modern recharge (i.e., over the last century where climate and soil data are available). Recharge estimation methods operating over modern timescales tend to be more easily impacted by land-use change. For example, Scanlon et al. (2006) demonstrate groundwater recharge both pre-and post-clearing in an Australian context, showing significant change (increase) in recharge.

RC7: So, even if I found this study very relevant and promising, I think that (1) the comparison with other product should be done more in depth, (2) a discussion about the salt-affected soils over Australia is perhaps relevant, and (3) a discussion about the impact of anthropogenic processes on groundwater chloride concentration (and then on your results) must be emphasized.

AC: No (additional) change suggested. We have addressed this comment in above responses.

Additional references not already included in the manuscript

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United Nations Environment Programme: World Atlas of Desertification: Second Edition, 1997.