

1 This paper presents a global data set of surface air concentrations and depositional
2 fluxes of ^7Be and ^{210}Pb that the authors compiled from literatures published during
3 1955-2020. This effort is timely as it has been a long time since last time such a data
4 set was compiled. The two radionuclides are very useful tracers for studying Earth's
5 surface (land/ocean) processes as well as transport and deposition processes in the
6 atmosphere. The new data set is expected to be widely used and cited in the years to
7 come. The content of this paper is generally well presented, but I do have some concerns
8 that should be addressed before its publication on ESSD.

9
10 We would like to thank the anonymous referee #2 for taking the time to provide a
11 thorough review of our submitted manuscript. The comments are very valuable and the
12 suggestions are very helpful. These comments and suggestions help us in greatly
13 improving the quality of our MS.

14 Below, the original comments are in black, our responses are in blue.

15
16 Major comments:

17
18 (1). There are many typos and grammatical errors in the text. Some are listed below.
19 Editing assistance is needed (perhaps from coauthor MB) and would significantly
20 improve the presentation.

21 Response: Thank you very much for pointing out the typos and grammatical errors in
22 the manuscript. We have corrected these typos and grammatical errors. Language has
23 been carefully further edited by one of the coauthors MB. Besides, the editorial team
24 of the ESSD will also edit the language if the manuscript is accepted, as presented in
25 the submission guidelines in the homepage of the journal.

26
27 (2). "Finally, we acknowledge that the seasonal information is indeed not much
28 discussed for this dataset. (P22, L456-457)"; "Further compilation of monthly data is
29 also warranted to assess seasonal variability of ^7Be and ^{210}Pb and understand the
30 relationship between these changes and influencing factors such as atmospheric
31 dynamics, meteorological condition, and geographic location on a global scale. (P23,
32 L465-468)"

33 ---- As authors mentioned in the paper, seasonal air concentrations and depositional
34 fluxes of ^7Be and ^{210}Pb are not reported. Such data would otherwise significantly
35 increase the value of this new compilation. For example, the seasonal data can be used
36 to evaluate seasonality of transport in global atmospheric models. The authors are
37 strongly encouraged to add the seasonal data into their data set, if at all possible. If not,
38 a discussion of why the seasonal data are not included would be helpful. In that case,
39 compiling the seasonal data in a future effort is also encouraged.

40 Response:

41 We totally agree that seasonal data would significantly increase the value of this new
42 dataset. Actually, seasonal ^7Be and ^{210}Pb data has never been compiled on a global scale.
43 We did try to compile some seasonal data of ^7Be and ^{210}Pb , but this completion work is
44 incomplete. Most of the data for seasonal studies are presented in graphs not in tables,

and in many older papers, the quality of graph and the paper used is poor and have to compromise the precision in extracting the data. Second, in some papers, although seasonal data were measured, only the annual data were provided. Furthermore, wherever there are seasonal data, it is important to have data on the amount of precipitation along with radionuclide data, as seasonal variations on the amount of precipitation plays a major role on the atmospheric scavenging and their depositional flux. Last and most importantly, since we were unable to retrieve reliable data from the graphs/charts, we reached out to some of the original authors for their original data, but received little help. And many of the older references, the authors no more active with their research and/or have retired or no more alive. Due to these constraints, we currently only have compiled only partial seasonal data, which is far from our ultimate goal. Many funding agencies now require that researchers submit their data to a public domain (such as National Science Foundation in USA, GEOTRACES Program) which will be accessible to global scientific community. More funding agencies should encourage to either join such efforts or start one in their home country and such data must be available for global scientific community, with no strings attached. We plan to reach out researchers who have still access to their seasonal data and try our best to compile the seasonal data in a future effort (may be need 1-2 y), then update the current version of the dataset.

In addition to the constraints listed above, adding seasonal data and related discussions will likely make this paper too lengthy, and thus have focused on annual data in the current work. To alleviate the concerns of the reviewer, we have added a short paragraph (given below) at the end of section 3.7 giving the rationale why we have not included seasonal data.

“Finally, we acknowledge that the seasonal data of ^7Be and ^{210}Pb has not been included in the current version of dataset, because compiling the seasonal data is more challenging than compiling the annual data. Unlike the annual data, most of the published seasonal data are presented in graphs, without giving in tables, and in some cases, the graph quality was poor and precision in data extraction is expected to be poor. Besides, in some papers, although seasonal data were measured, only the annual data were provided. Thus, the comprehensive compilation of seasonal data of ^7Be and ^{210}Pb may need collaboration with and data sharing from the scientific community. The compilation of seasonal data is expected to be useful to assess seasonal variability of ^7Be and ^{210}Pb and understand the relationship between these changes and influencing factors such as atmospheric dynamics, meteorological conditions, and geographic location on a global scale. And the seasonal data can also be useful in evaluating seasonality of transport in global atmospheric models.”

Because there is no discussion on seasonal variations, the title of this paper is now changed to “A global dataset of atmospheric ^7Be and ^{210}Pb measurements: **annual** air concentration and depositional flux”

(3). Is the unit of air concentration “ mBq m^{-3} ” or “ mBq / SCM ” where “SCM” stands for standard cubic meter?

Response: The unit of air concentration is “ mBq m^{-3} ”.

(4). P2, L34-35: “Depositional flux of ^7Be is independent of longitude but depends on the altitude and the ~ 11 years solar cycle”

As Figure 4c shows, the ^7Be depositional flux does depend on longitude, and the error bars show the longitudinal variability of ^7Be deposition fluxes is quite large at northern mid-latitudes. Do you mean the production rate of ^7Be is independent of longitude? Do you mean “latitude” by “altitude” here?

Response: Thank you for noting the mistake here. Here we originally intended to express that the production rate of ^7Be is independent of longitude. And the word “latitude” was missed here. This sentence is now rewritten as “**The production rate of ^7Be has negligible dependence on longitude or season, but depends on altitude, latitude and the ~ 11 years solar cycle (Koch et al., 1996; Liu et al., 2001; Su et al., 2003)**”. And to make the text more coherent, this sentence will be moved forward at the end of the sentence “ ^7Be , a cosmogenic radionuclide, is produced by the spallation of oxygen and nitrogen nuclei by cosmic rays in the stratosphere and upper troposphere.”

Reference:

Koch, D. M., Jacob, D. J., and Graustein, W. C.: Vertical transport of tropospheric aerosols as indicated by and in a chemical tracer model, *J. Geophys. Res.*, 101, 18651-18618, 1996.

Liu, H., Jacob, D. J., Hey, I., and Yantosca, R. M.: Constraints from ^{210}Pb and ^7Be on wet deposition and transport in a global three-dimensional chemical tracer model driven by assimilated meteorological fields, *J. Geophys. Res.*, 106, 12109-12128, 2001.

Su, C. C., Huh, C. A., and Lin, F. J.: Factors controlling atmospheric fluxes of ^7Be and ^{210}Pb in northern Taiwan, *Geophys. Res. Lett.*, 30, <https://doi.org/10.1029/2003GL018221>, 2003.

P22, L441-444: “As mentioned above, ^7Be depositional flux is independent of longitude and is constant over latitudinal bands. Thus, the ^7Be depositional flux data in our dataset can be used to estimate ^7Be ocean inventory in the same latitude, which can avoid the collection of the large volume of seawater samples and extend the application of ^7Be in the Open Ocean.”

Again, see the comment above. In that case, the ^7Be depositional flux data in the dataset would not be able to be used to estimate ^7Be ocean inventory in the same latitude.

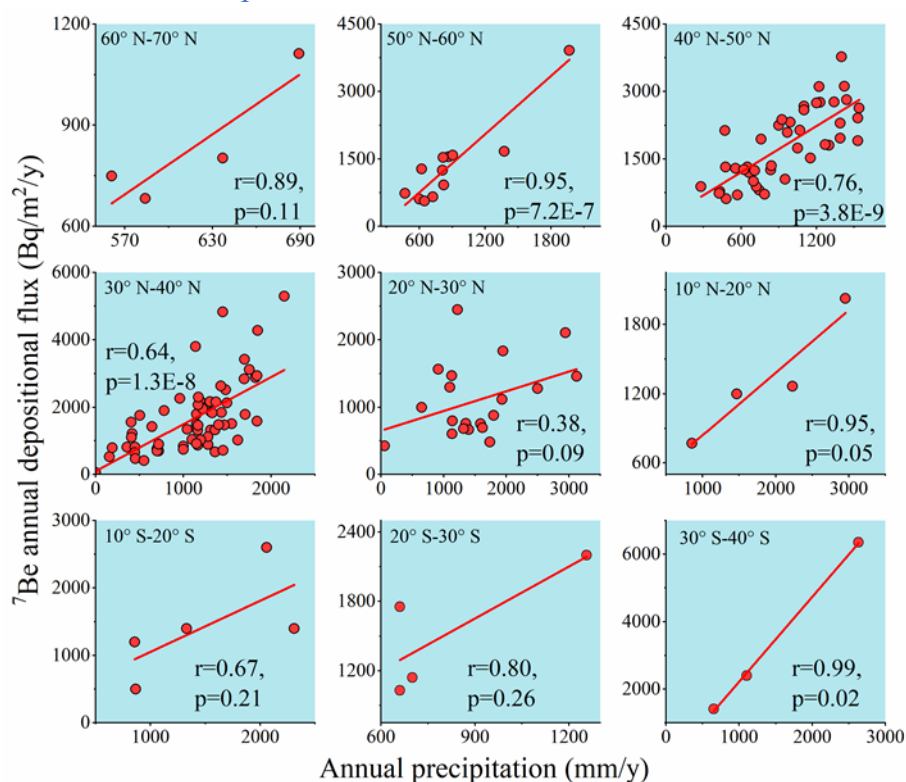
Response:

Indeed, in Figure 4c, the ^7Be depositional flux varies with longitude even within the specific latitudinal bands, but we believe **such variability is mainly due to spatial variations in the amount of precipitation** since ^7Be is removed from atmosphere primarily by precipitation. The dataset supports this observation. As shown in Fig. 7 (see below), **^7Be annual depositional fluxes generally show a significant positive correlation with annual amount precipitation**, especially at the northern mid-latitudes where the data coverage is good. In this case, the empirical equation between ^7Be depositional fluxes and annual precipitation provide an empirical method for estimating fluxes, although frequency of precipitation also is likely a factor.

To alleviate the concerns of this reviewer and the other reviewer, we have added a new paragraph (as below) outlining and clarifying the use of the dataset. Besides, the empirical equations describing the relationships between annual precipitation and ^7Be depositional fluxes for different latitudinal belts is also added as a new table in the

revised manuscript.

“Our dataset provides a forum in which a large amount of ^7Be and ^{210}Pb atmospheric depositional flux data for the above-mentioned research communities. This database will help in identifying data gaps and evaluating the empirical relations between ^7Be and ^{210}Pb depositional fluxes and annual precipitation. Researchers can rely on previously collected data in planning their research, without additional monitoring of ^7Be and/or ^{210}Pb depositional fluxes. Even for those areas with data gaps, the empirical equations between ^7Be and ^{210}Pb depositional fluxes and annual precipitation provide an empirical method for estimating fluxes, especially for ^7Be , as ^7Be depositional flux is independent of longitude and is constant over broad latitudinal bands. In summary, the atmospheric depositional flux data presented in our dataset along with the meta-analysis of the data will be useful in the investigations of soil erosion studies in terrestrial environments, particle dynamics studies in aquatic systems, and surface mixing process studies in open ocean.”



Minor comments:

P1, L29: Earth’s surface AND ATMOSPHERIC processes

Response: Thank you for the suggestion. “and atmospheric” will be added here in the revised manuscript.

P2, L32-34: correct grammar.

Response: Thank you for noting this mistake. This sentence is rephrased as “A major fraction of ^7Be (67%) production takes place in the stratosphere, but it does not readily reach the troposphere except during spring when seasonal thinning of tropopause folds

near the jet stream take occurs at mid-latitudes (Lal and Peters, 1967; Danielsen, 1968). Thus, ^7Be flux to the Earth's surface varies with latitude and season (Lal and Peters, 1967; Koch and Mann, 1996)."

Reference

Danielsen, E. F.: Stratospheric-tropospheric exchange based on radioactivity, ozone, and potential vorticity. *J. Atmos. Sci.*, 25, 502-518, 1968.

Koch, D. M. and Mann, M. E.: Spatial and temporal variability of ^7Be surface concentration, *Tellus B*, 48, 387-396, 1996.

Lal, D. and Peters, B.: Cosmic ray produced radioactivity on the Earth, in: *Handbuch der Physik / Encyclopedia of Physics*, edited by: Sittle, K., Springer, Berlin, Heidelberg, Germany, 551-612, https://doi.org/10.1007/978-3-642-46079-1_7, 1967.

P2, L56: studyING

Response: Thank you for noting this mistake – it is corrected in the revised manuscript.

P2, L56: add comma after all "e.g." throughout the text

Response: Thank you for noting this mistake – we have added comma after all "e.g." in the revised manuscript.

P3, L80: fluxes OF ^7Be

Response: Thank you for noting this mistake – it is corrected in the revised manuscript.

P3, L84: "To date, only one dataset was published that compiled ^7Be and ^{210}Pb together (Persson, 2016)" --- is it actually a 2015 publication?

Persson, B. R. R. (2015) Global distribution of ^7Be , ^{210}Pb and ^{210}Po in the surface air. *Acta Scientiarum Lundensia*, Vol.2015-008, pp.1-24. ISSN 1651-5013

Response: Thank you for noting this mistake – it is corrected in the revised manuscript. The corresponding reference in the reference list is also corrected.

P4, L111: This is confusing. Correct grammar. Complementary is an adjective.

Response: Thank you for noting this mistake. This sentence is now rephrased as "using natural archives avoids the labor and time-intensive measurements of ^7Be and ^{210}Pb concentration in precipitation and can serve as a complement to ..."

P5, L133: Alternately - do you actually mean "Alternatively, "

Response: Thank you for noting this mistake – it is corrected in the revised manuscript.

P6, L158-162: This sentence is way too long and hard to understand. Please revise.

Response: Thank you for the suggestion. This sentence is now split into two sentences: "It is expected that the ^7Be inventory is season-dependent in areas with large seasonal variations in precipitation (e.g., monsoon-dominated continental and oceanic areas). Time-series study in Bermuda has shown that the inventory of ^7Be was relatively

constant throughout the year, such that ^7Be inventory measured at any one time is likely representative (to within 20%) of the instantaneous ^7Be flux (Kadko et al., 2015)."

Reference

Kadko, D., Landing, W. M., and Shelley, R. U.: A novel tracer technique to quantify the atmospheric flux of trace elements to remote ocean regions, *J. Geophys. Res-Oceans*, 120, 848-858, 2015.

P6, L171: "and hence those are data are not included" – please rewrite.

Response: Thank you for the suggestion. This sentence is rewritten as "the data of ^7Be soil inventory are not included in our dataset".

P7, L175: , AND the latter

Response: Thank you for noting this mistake – it is corrected in the revised manuscript.

P7, L200: remove "the" before "dating ice core"

Response: Thank you for noting this mistake – it is removed in the revised manuscript.

P7, L204: typo "filed" (field)

Response: Thank you for noting this mistake –it is corrected in the revised manuscript.

P8, L209: can ALSO be obtained

Response: Thank you for noting this mistake –it is corrected in the revised manuscript.

A similar mistake is also corrected.

P8, L223: only those sites WITH more than one year of data

Response: Thank you for noting this mistake – it is corrected in the revised manuscript.

P10, L255: THE number of

Response: Thank you for noting this mistake –it is corrected in the revised manuscript.

P13, L301: "a sharp increment in ^7Be air concentration occurred on the Antarctic continent" – this reflects the subsiding motion of air over the Antarctic continent

Response: Thank you for the suggestion. This sentence will be rewritten as "a sharp increment in ^7Be air concentration (lack of flux data) occurred on the Antarctic, which reflects the subsidence of stratospheric air masses over the Antarctica continent (Wagenbach et al., 1988; Elsässer et al., 2011)."

Reference

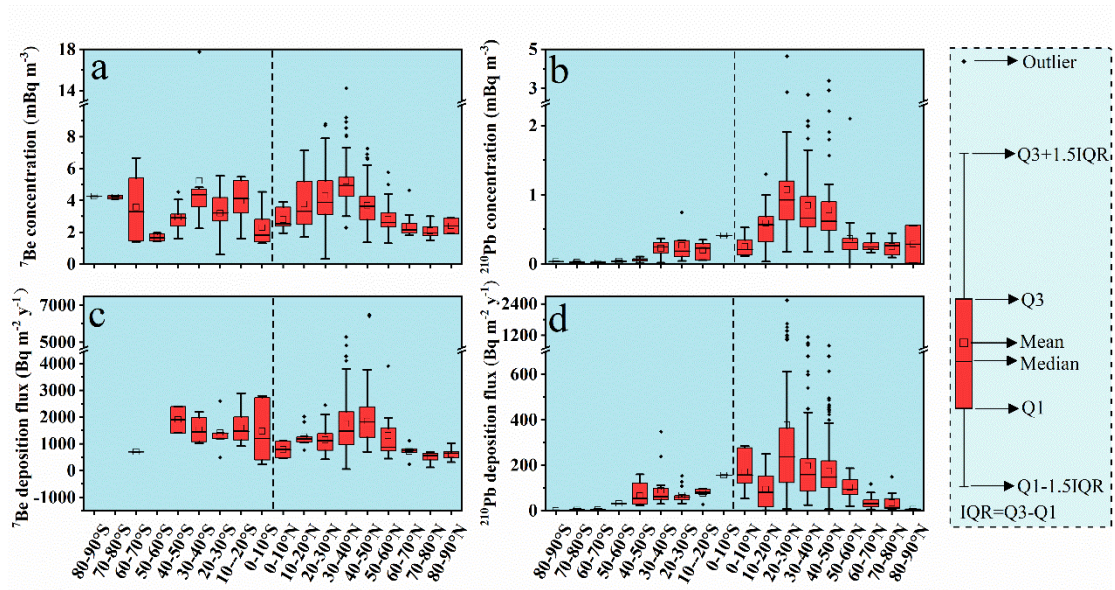
Elsässer, C., Wagenbach, D., Weller, R., Auer, M., Wallner, A., and Christl, M.: Continuous 25-yr aerosol records at coastal Antarctica, *Tellus B*, 63, 920-934, 2011.

Wagenbach, D., Görlach, U., Moser, K., and Münnich, K. O.: Coastal Antarctic aerosol: the seasonal pattern of its chemical composition and radionuclide content, *Tellus*, 40B, 426-436, 1988.

P15, Fig.5: the convention is to plot from South to North (x-axis). Also indicate what the whiskers / dots / bars stand for.

Response: The Fig. 5 has been replotted as below, and have added a legend to indicate

what the whiskers / dots / bars stand for.



P21, L406: “CTM” is the abbreviation for chemical transport model; it’s not a model name.

How about “a CTM based on GISS GCM”?

Response: Thank you for the suggestion. “CTM” will be changed as “CTM based on GISS GCM”. In addition, the model “GMI CTM” (Liu et al., 2016) is also added in this sentence.

Reference

Liu, H., Considine, D. B., Horowitz, L. W., Crawford, J. H., Rodriguez, J. M., Strahan, S. E., Damon, M. R., Steenrod, S. D., Xu, X., Kouatchou, J., Carouge, C., and Yantosca, R. M.: Using beryllium-7 to assess cross-tropopause transport in global models, *Atmos. Chem. Phys.*, 16, 4641-4659, 2016.

P21, L431-432: Not sure what “Bq m-2 y-1 / mean-life of the isotope, y)” means.

Response: Based on the suggestion of anonymous referee #1, we have made major revisions (as below) of section 3.6, and this sentence has been deleted in the revised manuscript.

The sentence in L428-436 is rewritten as “In aquatic systems (including river, lake, estuary and coast), the mass balance models of ^7Be and $^{210}\text{Pb}_{\text{ex}}$ have become powerful tools to understand the sediment source, transportation and resuspension processes (e.g. Wieland et al., 1991; Feng et al., 1999; Jweda et al., 2008; Huang et al., 2013; Mudbidre et al., 2014). In such models, the atmospheric depositional input of ^7Be and ^{210}Pb is a required source term. In addition, $^7\text{Be}/^{210}\text{Pb}_{\text{ex}}$ activity ratio can be used to identify the source area of sediments (Whiting et al., 2005; Jweda et al., 2008; Wang et al., 2021), to quantify the age of sediments (Matisoff et al., 2005; Saari et al., 2010), and to determine the transport distance of suspended particles (Bonniwell et al., 1999, Matisoff et al., 2002). Thus, the atmospheric depositional flux data of ^7Be and ^{210}Pb are also important for tracing particle dynamics in aquatic systems’

Reference

- Bonniwell, E. C., Matisoff, G., and Whiting, P. J.: Determining the times and distances of particle transit in a mountain stream using fallout radionuclides, *Geomorphology*, 27, 75-92, 1999.
- Feng, H., Cochran, J. K., and Hirschberg, D. J.: ^{234}Th and ^7Be as tracers for the transport and dynamics of suspended particles in a partially mixed estuary, *Geochim. Cosmochim. Ac.*, 63, 2487-2505, 1999.
- Huang, D., Du, J., Moore, W. S., and Zhang, J.: Particle dynamics of the Changjiang Estuary and adjacent coastal region determined by natural particle-reactive radionuclides (^7Be , ^{210}Pb , and ^{234}Th), *J. Geophys. Res-Oceans*, 118, 1736-1748, 2013.
- Jweda, J., Baskaran, M., van Hees, E., and Schweitzer, L.: Short-lived radionuclides (^7Be and ^{210}Pb) as tracers of particle dynamics in a river system in southeast Michigan, *Limnology and Oceanography*, 53, 1934-1944, 2008.
- Matisoff, G., Bonniwell, E. C., and Whiting, P. J.: Radionuclides as Indicators of Sediment Transport in Agricultural Watersheds that Drain to Lake Erie, *Journal of Environmental Quality*, 31, 62-72, 2002.
- Matisoff, G., Wilson, C. G., and Whiting, P. J.: The $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$ ratio as an indicator of suspended sediment age or fraction new sediment in suspension, *Earth Surf. Proc. Land.*, 30, 1191-1201, 2005.
- Mudbidre, R., Baskaran, M., and Schweitzer, L.: Investigations of the partitioning and residence times of Po-210 and Pb-210 in a riverine system in Southeast Michigan USA. *J. Environ. Radioact.*, 138, 375-383, 2014.
- Saari, H. K., Schmidt, S., Castaing, P., Blanc, G., Sautour, B., Masson, O., and Cochran, J. K.: The particulate $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$ and $^{234}\text{Th}/^{210}\text{Pb}_{\text{xs}}$ activity ratios as tracers for tidal-to-seasonal particle dynamics in the Gironde estuary (France): implications for the budget of particle-associated contaminants, *Sci. Total. Environ.*, 408, 4784-4794, 2010.
- Wang, J., Du, J., Baskaran, M., and Zhang, J.: Mobile mud dynamics in the East China Sea elucidated using ^{210}Pb , ^{137}Cs , ^7Be , and ^{234}Th as tracers, *J. Geophys. Res-Oceans*, 121, 224-239, 2016.
- Wang, J., Huang, D., Xie, W., He, Q., and Du, J.: Particle Dynamics in a Managed Navigation Channel Under Different Tidal Conditions as Determined Using Multiple Radionuclide Tracers, *J. Geophys. Res-Oceans*, 126, e2020JC016683, 2021.
- Whiting, P. J., Matisoff, G., Fornes, W., and Soster, F. M.: Suspended sediment sources and transport distances in the Yellowstone River basin, *Geol. Soc. Am. Bull.*, 117, 515-529, 2005.
- Wieland, E., Santschi, P. H., and Beer, J.: A multitracer study of radionuclides in Lake Zurich, Switzerland: 2. Residence times, removal processes, and sediment focusing, *J. Geophys. Res-Oceans*, 96, 17067-17080, 1991.

P22, L450: change “in areas” to “areas”

Response: The suggestion will be taken in the revised manuscript. “Concerning air concentrations in areas such as...” will be changed as “Concerning air concentrations, areas such as...”

P22, L454: “which ARE almost”

Response: Thank you for noting this mistake – it is corrected in the revised manuscript.

P23, L470: correct “SO4-“.

Response: Thank you for noting this mistake –it is corrected in the revised manuscript.

320
321 P23, L471-472: what is the connection between the 1st and 2nd sentences?

322 Response: Thank you for the suggestion. In order to make the text more connected and
323 coherent, we have reorganized this paragraph (as below, move 1st sentence forward)
324 "... quantification of the role of dry fallout in the removal of these nuclides will provide
325 insights on the removal of other analog species. As mentioned earlier, combining
326 cosmogenic ^7Be with ^{210}Pb which has a predominantly Earth-surface origin will be
327 useful to trace species that originate both from Earth's surface, such as Hg , SO_4^{2-} , NO_3^- ,
328 and those that originate in the upper atmosphere, such as O_3 . The size distribution of
329 aerosols particles carrying ^7Be and ^{210}Pb is crucial for understanding atmospheric
330 behavior and tracing analogues, and such studies also need to be conducted. Besides,
331 the troposphere contains ~99% of global water vapor with < 1% in the stratosphere.
332 The depositional velocity of aerosol in the stratosphere... "

333
334 P23, L473-474: how about zonal transport?

335 Response: Thank you for the suggestion. This sentence is rephrased as "the ^7Be
336 concentration is governed by local production, zonal and vertical downward transport,
337 and its decay".

338
339 P23, L474-477: Do these lines mean the following? "In the middle and upper
340 troposphere where precipitation is much less frequent, the removal rate of aerosols is
341 also slow. Collection of air samples in that part of the atmosphere will provide useful
342 information on the total deposition velocity of aerosols (Lal and Baskaran, 2012)."

343 Response: Yes, our meaning here is consistent with the sentences you wrote above. We
344 will replace these lines with the above sentences in the revised manuscript.
345