

## **Responses to RC1**

We thank the reviewer for the thoughtful comments and suggestions. Our responses are in blue text. The line and page numbers in our responses refer to those in the revised manuscript with track changes.

General Comments:

The sensitivity of Rn-222 to emission data sets is examined using GEOS-Chem driven by meteorological fields from two sources.

This manuscript would be stronger and of more scientific significance if it used MERRA-2 meteorological fields, which have been available for several years, and if the simulations were performed at  $1 \times 1.25^\circ$ , which has become the standard resolution for global simulations.

We generally agree that our work would be more scientifically worthwhile if we could use the MERRA-2 reanalysis. However, MERRA was the most widely used reanalysis when we first started this study. To compare with transport in the more recent version of GEOS data, we included in the paper the model simulation driven by GEOS-FP (“Forward Processing”), the operational GEOS meteorological data product.

We also agree that a higher resolution would improve the simulation of transport in the model. However, the classic GEOS-Chem is not available at a resolution higher than  $2 \times 2.5^\circ$  for global simulations. The High-Performance version of GEOS-Chem (GCHP) runs at much higher resolution (c180, equivalent to  $0.5 \times 0.625^\circ$ ) with a cubed sphere grid geometry. Rn-222 and Pb-210 simulations with GCHP will be tested and evaluated in future studies.

The phrase “excessive Asian emissions” is confusing. It could be misinterpreted as indicating that model emissions are too high over Asia. Please go through manuscript and rephrase as necessary.

The phrase “excessive Asian emissions” was used in the manuscript to describe that the actual emissions in Asia are likely significantly higher than prescribed emissions in the model. We have replaced “excessive Asian emission” with “underestimated emission in Asia” in a few places to avoid confusion: Line 12 on Page 6, Line 14 on Page 10, Line 2 on Page 20, and Line 13 on Page 32.

Perhaps a Table could be added that concisely summarizes differences between the 4 emission scenarios. It’s tedious reading through 4 pages describing the different emission scenarios. You correctly point out the deficiencies of using a simplistic emission inventory but you should also mention that there are benefits for using a simplistic emission inventory. For example, Rn-222 concentrations can be used as an indicator for how recently an air mass encountered land or a quick method of interpreting the effects of changes in the model configuration on convective mixing.

Thanks for the suggestion. We have added a new table (Table 1 in the revised manuscript and also given below) to summarize the differences between the four emission scenarios.

**Table 1. Global <sup>222</sup>Rn emission scenarios used in this work.**

Scenario	Reference	Description
JA97	Jacob et al. (1997)	Emission fluxes are 1.0 atom cm <sup>-2</sup> s <sup>-1</sup> over land between 60°N – 60°S, 0.005 atom cm <sup>-2</sup> s <sup>-1</sup> between 60°N – 70°N and 60°S – 70°S, zero poleward of 70°N/S, and 0.005 atom cm <sup>-2</sup> s <sup>-1</sup> over lakes and oceans. Emissions are reduced by a factor of 3 when surface temperature is below 0°C.
SW98	Schery and Wasiolek (1998)	Emission fluxes are formulated by using a theoretical diffusion model of porous soil with controlling factors of soil radium content, soil moisture, and surface temperature. Emission fluxes in SW98 were found to be overestimated and are reduced by a factor of 1.6 globally in this work (Koch et al., 2006; Zhang et al., 2011)
ZK11	Zhang et al. (2011)	Based on SW98, ZK11 updated emission fluxes over Europe, U.S., China, Australia, and oceanic regions according to more recent measurements.
ZKC	This work	ZKC increases emission fluxes in the geographical territory of China by a factor of 1.2 upon ZK11 and retrogresses to SW98 over U.S.

We agree with the reviewer that with a spatially uniform and simplistic emission, Rn-222 is a good tracer to assess continental influence and convective mixing. We have added the following statement in Line 15 on Page 8:

“Since the emission fluxes in JA97 are fairly uniform over land area, this simplistic emission scenario can be used to discern continental influence on air masses in global models and assess the effect of any changes in the model representation of convective mixing (Balkanski et al., 1992; Jacob et al., 1997).”

Our perspective about the value of using a realistic Rn-222 emission scenario is that model simulated Rn-222 concentrations can be better compared with actual observations, thus providing an observation-based evaluation of transport processes in models.

The method used to construct the ZKC inventory is a bit confusing. Does it consist of the SW98 inventory over North America and the ZK11 inventory elsewhere, with the latter multiplied by a factor of 1.2 over China? If yes, say this concisely and include the latitude/longitude range for the factor of 1.2 adjustment. Why is this adjustment applied to China and not southern China? If you want people to use this inventory rather than ZK11 you need to document it better – here.

A description of the modifications we made to the ZKC inventory is now given in the added Table 1 (also see above). The factor of 1.2 adjustment was applied to the geographical territory of China.

The adjustment of Rn-222 emission fluxes over China (not just southern China) is made based on the analyses of Rn-222 surface concentrations and Pb-210 deposition fluxes (Du et al., 2015). Some sites used in the Pb-210 deposition analysis are located in northern and inland China. We have clarified this in Line 6 on Page 20:

“We then calculate the correlations ... at the sites in North America (nine sites) and Asia (nine sites; Du et al. (2015)). Some studied Asian sites are located in northern and inland China.”

In general, the figures are of excellent quality and enlightening.

## Specific Comments

P1 L25 (add lower bound to <70%)

There is no lower bound. The “<70%” is an approximate fraction of data within the a-factor-of-2 range. To avoid confusion, we have replaced “<70%” by the exact value 68.9% in Fig. 6(m).

P5 L7-8 Please rephrase awkward sentence that begins with “Due to the availability of”

The original sentence reads:

“Due to the availability of extensive measurements of  $^{222}\text{Rn}$  emission fluxes and surface concentrations, Europe has the finest resolution emission inventory of up to  $0.083^\circ \times 0.083^\circ$  with variability in regional and temporal emissions”

We have rewritten the sentence as the following:

“Published  $^{222}\text{Rn}$  emission inventories for Europe have very fine spatial resolutions (up to  $0.083^\circ \times 0.083^\circ$ ) with monthly variability due to extensive measurements of emission fluxes and surface concentrations across the continent.”

P6 L19: TPCORE is internal nomenclature; please choose a more descriptive term such as monotonic if appropriate.

We have rewritten the sentence as the following:

“The model uses a flux-form semi-Lagrangian finite volume scheme, known as TPCORE, to calculate advection (Lin and Rood, 1996). The scheme uses the monotonic piecewise parabolic method under convergence conditions and a semi-Lagrangian method otherwise.”

P7: What is the difference between GEOS-FP and MERRA-2?

GEOS-FP is the current operational product of GEOS-5 and includes recent developments of the model. MERRA and MERRA-2 are long-term reanalysis products based on different versions of GEOS-5. We have revised the relevant sentence to “MERRA-2, which is based on a newer version of GEOS-5 and shows improved climate over MERRA (Molod et al., 2015), is not used here ....”.

P9 L25: What is your rationale for not including these updates?

Zhang et al. (2011) showed that Rn-222 emissions in EU calculated based on gamma-dose rate (Szegvary et al., 2009) lead to satisfactory agreement between model simulated and observed Rn-222 surface concentrations. Both López-Coto et al. (2013) and Karstens et al. (2015) used a different type of method. They used soil conditions to estimate Rn-222 emission fluxes and the uncertainties are largely determined by soil moisture in land surface models. Considering our second goal of assessing convection in GEOS-Chem and a better comparison with the results in Zhang et al. (2011), we chose to follow the method and analysis in Zhang et al. (2011). We have revised the statement in Line 16 on Page 10 to the following:

“Since ZK11 has been tested with satisfactory agreements between modeled and observed surface concentrations in Europe (Zhang et al., 2011), the updates for emission fluxes in Europe by López-Coto et al. (2013) and Karstens et al. (2015) are not included.”

P10 L7 Define Gucci

Added in the text “GCI (Giga-Curie)”.

P12 L4: How many sites are located in the SH?

Added in the text “Fewer sites (11) are located in the Southern Hemisphere”.

P12 L6: What do you mean by the Rn-222 observations were made in consecutive years. Is this true of all 51 sites? Do you mean at least 2 years of consecutive data are available at each site?

We have changed “consecutive years” to “multiple years”, as the measurements were not always consecutive.

P12: Any thoughts on why Rn-222 profile data sets are scarce given the proliferation of other profile data sets?

Thanks for bringing this up. We have added one sentence in the text: “The scarcity of <sup>222</sup>Rn airborne measurements is partly due to the fact that the measurement requires an extraction and counting facility nearby in order to minimize decay and that the process of radon extraction is labor-intensive (Williams et al., 2011).”

P13: Would make for a more interesting read if “significantly”, “substantially”, and “remarkably” were quantified.

We have quantified the changes in surface concentration differences in Line 12-13 and in Line 22-23 on Page 14.

P16 L2: Why would the annual means be less representative? Were they obtained from only a portion of the year?

Some annual means at China sites were obtained from measurements over the period of Nov. 1988 - Jan. 1990. The measurements only covered a little over a year and were less representative for the surface climatology compared to other sites where monthly measurements were reported for multiple years. We have added the following in the earlier data section:

“The few inland sites in China only reported annual means based on measurements of 1-2 years (Jin et al., 1998).”

Jin, Y., Iida, T., Wang, Z., Ikebe, Y., and Abe, S.: A subnationwide survey of outdoor and indoor <sup>222</sup>Rn concentrations in China by passive method. Radon and thoron in the human environment, in: Radon and Thoron in the Human Environment, in: Proceedings of the 7th Tohwa University International Symposium, edited by: Katase, A. and Shimo, M., World Scientific Publishing Co. Pre. Ltd., Singapore, 276–281, 1998.

P19 L1-4: This is a bit confusing. Increasing the scaling factor over China from 1.2 to a higher value would lead to a better agreement between simulated and observed deposition fluxes over Asia (Figure 7c).

However, this would also lead to an overestimate of deposition fluxes in the Northern Hemisphere. Please explain more clearly and/or reference the latter result.

We are working on a follow-up manuscript to compare model results with Pb-210 observations, including deposition fluxes and vertical profiles. The unpublished results indicate that larger scaling factors will lead to large overestimates over the rest of the Northern Hemisphere. We have changed the sentence to “...we choose to use a moderate scaling factor of only 1.2 for China to avoid large overestimates of total <sup>210</sup>Pb

deposition fluxes over the rest of the Northern Hemisphere”. The reference of the on-going Pb-210 work has been given in Line 4-5 on Page 17.

P21 L3: Please quantify the model high-bias.

We have added quantifications of model high biases here:

“Simulations with JA97 and SW98 overestimate the observations by a factor of >2 on average, while such large overestimates are only seen in February for ZK11 and ZKC.”

P25 L1: Are profiles of any more widely sampled trace gases useful for evaluating the convective detrainment level? I worry that some of the issue is with the observed profiles.

Vertical distribution of CO has been frequently used to examine convective transport in atmospheric models (e.g., Allen et al., 1996; Ott et al., 2009). Stanfield et al. (2019) found that the frequency distribution of the convective entrainment rates (mixing between environmental air with in-cloud air) for deep convection events in GEOS-5 has a significantly larger fraction in the higher-end values compared to the rates derived from TES/MLS-observed CO profiles. Intensive mixing within convective updraft undermines the upward lifting of surface air masses to the upper troposphere, possibly causing the rapidly decreasing  $^{222}\text{Rn}$  concentrations with height in the simulation with GEOS-FP (Figure 12a). The cloud-top height for convective clouds in MERRA is likely biased high according to a comparison with CERES-observed clouds (Posselt et al., 2012). These are consistent with our conclusions about the detrainment level derived based on  $^{222}\text{Rn}$  profiles. We have revised this part of the paragraph to the following to support our viewpoint:

“... MERRA exhibits a higher and deeper convection from 5 to 10 km. As a result, a remarkable underestimation of  $^{222}\text{Rn}$  concentrations with MERRA is seen from 4 to 8 km, followed by overestimations above 9 km. Deep convective cloud top in MERRA has been shown biased high compared to CERES-observed clouds (Posselt et al., 2012). Stanfield et al. (2019) found that the frequency distribution of convective entrainment rates (mixing between environmental air with in-cloud air) for deep convection events in GEOS-5 has a significantly larger fraction in the higher-end values compared to the rates derived from TES/MLS-observed CO profiles. Intensive mixing during convective updraft undermines the upward lifting of surface air masses to the upper troposphere, possibly causing the rapidly decreasing  $^{222}\text{Rn}$  concentrations with height in the simulation with GEOS-FP. Due to weaker convection in GEOS-FP, the simulation underestimates in a broader altitude range (4-10 km). It seems challenging for the two GEOS products to capture the convective detrainment level. ...”

Allen, D. J., Kasibhatla, P., Thompson, A. M., Rood, R. B., Doddridge, B. G., Pickering, K. E., ... & Lin, S. J. (1996). Transport-induced interannual variability of carbon monoxide determined using a chemistry and transport model. *Journal of Geophysical Research: Atmospheres*, 101(D22), 28655-28669.

Ott, L. E., Bacmeister, J., Pawson, S., Pickering, K., Stenchikov, G., Suarez, M., ... & Xueref-Remy, I. (2009). Analysis of convective transport and parameter sensitivity in a single column version of the Goddard earth observation system, version 5, general circulation model. *Journal of the atmospheric sciences*, 66(3), 627-646.

Posselt, D. J., Jongeward, A. R., Hsu, C. Y., & Potter, G. L. (2012). Object-based evaluation of MERRA cloud physical properties and radiative fluxes during the 1998 El Niño–La Niña transition. *Journal of climate*, 25(21), 7313-7327.

Stanfield, R. E., Su, H., Jiang, J. H., Freitas, S. R., Molod, A. M., Luo, Z. J., ... & Luo, M. (2019). Convective entrainment rates estimated from Aura CO and CloudSat/CALIPSO observations and comparison with GEOS-5. *Journal of Geophysical Research: Atmospheres*, 124(17-18), 9796-9807.

P30L15: Why would re-mapping have a greater impact on GEOS-FP than MERRA?

Re-mapping (from the cubed-sphere to equally rectilinear grids) itself does not have different impacts on GEOS-FP than MERRA. Convection in the simulation driven by GEOS-FP is affected more by a more intensive regridding from a finer native model resolution (0.25° by 0.3125° to 2° by 2.5°) compared to MERRA (0.5° by 0.667° to 2° by 2.5°). We added such information in an earlier place in Line 24 on Page 28:

“GEOS-FP has a finer native horizontal resolution (0.25° latitude by 0.3125° longitude) than MERRA reanalysis (0.5° latitude by 0.667° longitude), and exhibits weaker convection likely due to a more intensive regridding.”

We also revised the sentence in Line 5 on Page 33 to “The weak convection in GEOS-FP leads to large low biases of <sup>222</sup>Rn in the mid-high troposphere”.

Figure 7b – be sure to indicate in caption that these are annual mean values of deposition fluxes. Also, be clear as to what each symbol shows and how many total there are. Is it 9 x 5?

Now we indicate in the caption that the values of deposition fluxes are annual means. The number of model simulations (5) and surface sites (9) have been stated in the figure caption already. There are 9 x 5 = 45 points in total.

Figure 8: Be clear that you are comparing an observed climatology from various years to a simulation of 2013. Are standard deviations of monthly means available at any of the sites? If yes, consider adding them.

We have changed the first sentence in the caption to:

“Comparison between observed <sup>222</sup>Rn climatological monthly means (black lines) and simulated monthly means in 2013 (color lines) at selected surface sites in Europe.”

Unfortunately, standard deviations are available for only a few sites. For those sites, multiple years of monthly data are available from the data repository submitted with this manuscript.

Figure 12a. How much trust should we put into the observed profile based on multiple sites. How many of the profiles extended into the upper troposphere and were those from a small subset of the total locations?

As stated earlier in the manuscript, the composite profile is compiled from summertime observations at 23 sites over the Northern Hemisphere mid-latitude continental regions. It should provide a decent measure of summertime vertical <sup>222</sup>Rn distribution over land. More than half of the profiles reach up to 6-12 km; the composite profile is thus not biased by a small subset of observations in the upper troposphere. Now we state in the Fig. 12 caption “...from 23 locations over the Northern Hemisphere continents (Liu et al., 1984),..... In panel (a), more than half of the observed profiles reach up to 6-12km”.

Figure 13. When examining the effects of convection on trace gas profiles, it seems odd to show annual mean plots. Perhaps you should just show the summer hemisphere.

This comment is addressed together with the next one.

Figure 14. Please relabel this plot. The percent contribution cannot be zero. Perhaps call it the Percent Change in annual zonal mean  $^{222}\text{Rn}$  due to convection

We agree with the suggestion that summertime results better reflect the effects of convection. We have replaced annual zonal means in Figures 13&14 with zonal means averaged over June-July-August. The conclusions from these analyses are still the same except that the effects of convection are more obvious in the Northern Hemisphere. The relevant pieces of text have been updated accordingly. The figure 14 caption has been revised to “Percentage changes in zonal mean  $^{222}\text{Rn}$  concentrations averaged over June-July-August due to convective transport in the GEOS-Chem simulations ....” (see below).

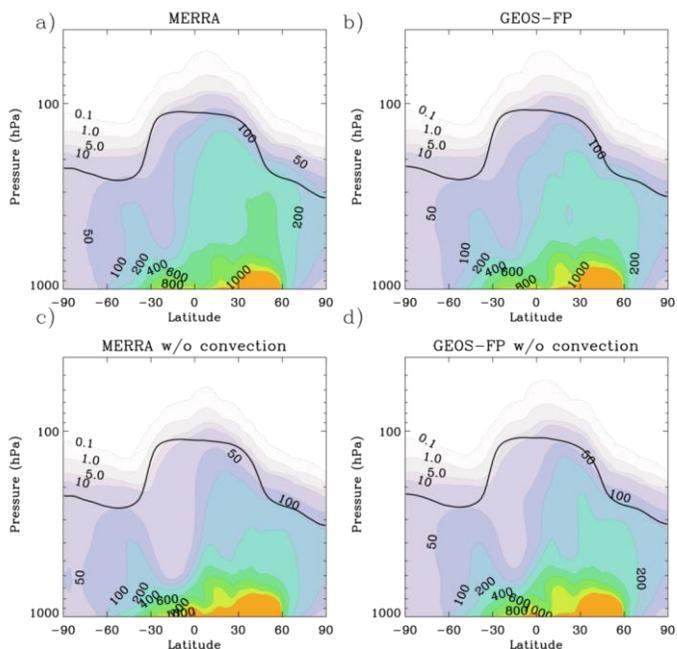


Figure 13. Latitude-pressure cross-sections of zonal mean  $^{222}\text{Rn}$  concentrations averaged over June-July-August (mBq/SCM) as simulated by the GEOS-Chem model driven by a) MERRA (A-1), b) GEOS-FP (B-1), c) MERRA without convection (A1-nc), and d) GEOS-FP without convection (B1-nc). Bold black lines denote the zonal mean tropopause height (hPa) in the corresponding meteorological data set.

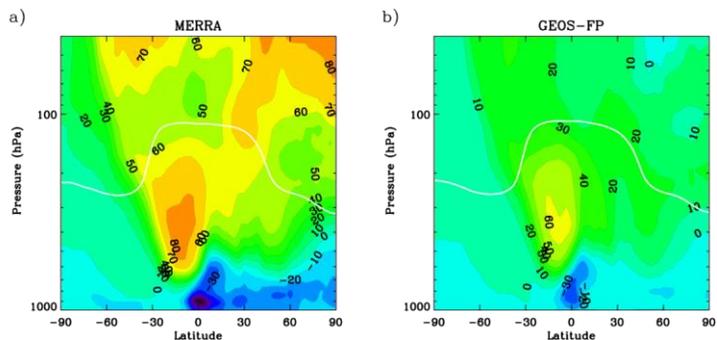


Figure 14. Percentage changes in zonal mean  $^{222}\text{Rn}$  concentrations averaged over June-July-August due to convective transport in the GEOS-Chem simulations driven by a) MERRA and b) GEOS-FP. Values are  $(^{222}\text{Rn} - ^{222}\text{Rn}_{\text{nc}})/^{222}\text{Rn} \times 100$ , where  $^{222}\text{Rn}$  and  $^{222}\text{Rn}_{\text{nc}}$  are  $^{222}\text{Rn}$  concentrations simulated with (A1 and B1, Table 1) and without (A1-nc and B1-nc) the convection operator, respectively.

**Technical Comments**

P3 L10 shaping 222Rn → shaping its

Done.

P3 L21 (remove period at end of line)

It is a comma not period. We will keep it there.

P3 L24 degree of discrepancies → discrepancies

P6 L3: apparent changes → changes

P7 L9: considerable → a considerable

P11 L5 further low latitudes → low latitudes

All done.

P11 L15: “changes are possible depending on the availability of measurements in these areas “ → changes are possible when measurements become available in these areas

P15L23: Replace excessive with large

P16 L19 Consider replacing “tentative” with “provisional”

P17 L11 three times at → three times higher at

All done.

P29 L22 & L24: Replace excessive with very large

This has been addressed in the second major comment above.

Figure 13: Replace GESO with GEOS.

P45 L8: range with → range within

Done.

## Responses to RC2

We thank the reviewer for the comments and insights. Our responses are in blue text. The line and page numbers in our responses refer to those in the revised manuscript with track changes.

As far as I am aware of, this study presents the most comprehensive piece of work to date using  $^{222}\text{Rn}$  to evaluate atmospheric transport and mixing on a global scale. It includes the assessment of four  $^{222}\text{Rn}$  emission scenarios, a CTM driven by two meteorological data sets, and the comparison of simulations with practically all atmospheric  $^{222}\text{Rn}$  observations currently available, including vertical profiles. The clear structure of the paper, its great readability and meaningful displays make it a pleasure to read. It leaves no open question to me. There is very little that I can suggest to further improve it.

### Minor comments

Differences between simulated and observed atmospheric concentrations occur for various reasons. One is the bias in measurement techniques, especially the underestimation of  $^{222}\text{Rn}$  concentrations derived from  $^{222}\text{Rn}$  progeny measurements near the surface (< 100 m above ground; cf. Grossi et al., 2020). Further,  $^{222}\text{Rn}$  concentration gradients within the first few metres above ground can be steep (e.g. Chambers et al., 2011). Several of the atmospheric observations in China were done between 1 and 1.5 m above ground (Jin et al., 1998, cited in Zhang et al, 2011, cited in the present study), which might explain some of the difference between simulation and observation for those sites. Are those sites represented in Figure 6 e-h by points indicating simulated values more than a factor of two smaller than observed values (or, better, observed values exceeding simulated values by more than a factor of two)?

The comparisons between observations and model results for Asian sites (Fig. 6e-h) suggest that surface  $^{222}\text{Rn}$  concentrations were underestimated by at least a factor of two in the model for a few sites. As the reviewer pointed out, the measurements have been taken very close to the surface. According to Figure 1 in Chambers et al. (2011),  $^{222}\text{Rn}$  concentrations measured close to surface can be significantly higher than those at 50m between 8pm until 9am, and are possibly higher than the average taken from the model bottom-layer gridbox (~100m high). On the other hand, there are possible low biases in the measurements due to measurement techniques. Such low biases may partially compensate for the underestimate due to the steep concentration gradients near the surface. Considering these rationales, we have added the following discussion in Line 16 on Page 16:

“The observations in China were taken between 1m and 1.5m above ground according to Jin et al. (1998). The model surface layer concentrations usually represent the averages in the model bottom layer (~100m high), and thus may be literally lower than the observations due to the steep concentration gradients near the surface, especially during nighttime (Chambers et al., 2011). On the other hand, there are possible low biases in the  $^{222}\text{Rn}$  concentrations derived from  $^{222}\text{Rn}$  progeny measurements (Schmithüsen et al., 2017; Grossi et al., 2020), lessening the above model underestimate due to large near-surface vertical gradients. These biases differ on a case-by-case basis and are difficult to quantify.”

Figure 6, y-axis label in the second row (Panel e) is "Observed ..." Should this not be "Simulated ...", as in the other rows?

Thanks for catching this typo. Now corrected.

Page 19, lines 7 and 8: "The seasonality in surface  $^{222}\text{Rn}$  concentrations is mainly affected by three factors: (1) the surface  $^{222}\text{Rn}$  emission flux rate determined by radium content and soil conditions; ..." This sentence is subject to eventual misinterpretation, in the way that radium content may be misunderstood as being seasonally variable. I would suggest to change the sentence to something like: "The seasonality in surface  $^{222}\text{Rn}$  concentrations is mainly affected by three factors: (1) seasonality in surface  $^{222}\text{Rn}$  emission flux rate resulting from seasonal changes in soil moisture, diffusivity, depth of the water table, snow and ice cover; ..."

Thanks for the suggestion. We have revised the sentence to:

"The seasonality in surface  $^{222}\text{Rn}$  concentrations is mainly affected by three factors: (1) the variability in  $^{222}\text{Rn}$  emission flux rate due to seasonal changes in soil moisture, diffusivity, depth of the water table, and snow and ice coverage; ..."

Page 24, lines 1 and 2: Some  $^{222}\text{Rn}$  flux measurements from Antarctic soil are reported in Evangelista and Pereira (2002). As mentioned in the text, there are vast regions without atmospheric  $^{222}\text{Rn}$  observations. Perhaps suggest, where from a modeller's perspective it would be desirable to see an atmospheric  $^{222}\text{Rn}$  detector established. Personally, I would very much like to see that happen at the tall tower (300 m) at Zotino (60 N 90 E), in the middle of Siberia (<http://www.zottoproject.org/index.php/Main/Home>).

We thank the reviewer for pointing us to this  $^{222}\text{Rn}$  flux measurement work in the Antarctic. The work provides valuable measurement of  $^{222}\text{Rn}$  fluxes during the summer of 1998/1999 at the Admiralty Bay area of King George Island, Antarctic Peninsula (62°S, 58°W). Reported fluxes ranged between  $0.21 \times 10^{-2}$  atom  $\text{cm}^{-2} \text{s}^{-1}$  and  $28 \times 10^{-2}$  atom  $\text{cm}^{-2} \text{s}^{-1}$ . We have added the following discussion in the text:

"Evangelista and Pereira (2002) reported summertime  $^{222}\text{Rn}$  fluxes ranging between  $0.21 \times 10^{-2}$  atom  $\text{cm}^{-2} \text{s}^{-1}$  and  $28 \times 10^{-2}$  atom  $\text{cm}^{-2} \text{s}^{-1}$  during the summer of 1998/1999 at the Admiralty Bay area of King George Island, Antarctic Peninsula (62°S, 58°W). The work also suggested such low fluxes could not explain  $^{222}\text{Rn}$  concentration surges in the atmosphere. The sparse measurements at the edge of the Antarctic are not adequate for inferring emission fluxes over the remote continent."

Regarding the desire for more observations, we have made a few statements in section 3.2 to suggest more measurements in Asia, North America, and Antarctic. Measurements in the middle Siberia would be very valuable because they may help quantify  $^{222}\text{Rn}$  emissions and surface concentrations in the northern Asia.

#### References

- Chambers et al. (2011) Separating remote fetch and local mixing influences on vertical radon measurements in the lower atmosphere. <https://doi.org/10.1111/j.1600-0889.2011.00565.x>
- Grossi et al. (2020) Intercomparison study of atmospheric  $^{222}\text{Rn}$  and  $^{222}\text{Rn}$  progeny monitors. <https://doi.org/10.5194/amt-13-2241-2020>
- Evangelista and Pereira (2002) Radon flux at King George Island, Antarctic Peninsula. [https://doi.org/10.1016/S0265-931X\(01\)00137-0](https://doi.org/10.1016/S0265-931X(01)00137-0)

### **Responses to Editor's comments**

We thank the editor for his questions. Our replies are in blue text.

There are two questions for which I did not find the answer in the manuscript:

1- What is the effect of the model resolution, i.e. would you get better agreement (or not?) with a  $0.1^\circ \times 0.1^\circ$  horizontal resolution?

As stated in our response to Review#1, we do think a higher resolution would improve the simulation of transport (i.e., minimizing lost vertical transport due to the regriding of meteorological fields) in the model. In the context of Rn-222 modeling, we doubt that model simulations with very fine resolutions can improve the scientific results of this work, largely due to the uncertainties and coarse resolution associated with Rn-222 emission maps. Studies of Rn-222 emission fluxes so far do not provide sufficient data required to produce fine-resolution emission maps, except for Europe. On the other hand, the classic version of the GEOS-Chem model is not available for conducting global simulations at a resolution higher than  $2^\circ \times 2.5^\circ$ . In future studies, Rn-222/Pb-210 simulations will be tested with GCHP, the High-Performance version of GEOS-Chem that can be run at high resolution with a cubed sphere geometry.

2- What is the effect of the geographical position of the  $^{222}\text{Rn}$  station in the model grid box when the station is on the coast such as for Fuzhou? Have you thought what would change in your simulated concentration if the model box was 80% marine / 20% continental versus 80% continental / 20% marine?

We appreciate Editor's insight that the observations and sampled model results may represent different air masses at coastal regions due to the nonuniform surface type in the model grid box. For coastal sites such as Fuzhou, the Rn-222 observations could be largely affected by local emissions and thus more typical of those at inland sites. Coastal sites are usually positioned within model gridboxes that are partly land and partly water. Some of these gridboxes may be dominated by water whereas the sites could be located near the edges of the gridboxes and not close to the water. In such cases, while the land area fractions (excluding sea and lake areas) are taken into account in the model calculation of Rn-222 emission fluxes for these gridboxes, the model-simulated concentrations may not represent the observed at the sites due to the subgrid nature of the latter.

We thus tried sampling the model results at adjacent gridboxes and found that the model results for the gridbox to the west of the original (moving away from coast) are much more comparable to the observed magnitude and seasonality of surface Rn-222 concentrations at Fuzhou. This suggests that the observations are significantly affected by local Rn-222 emissions. We also found a similar improvement at the site of Hong Kong. We replaced model results in Figure 9(c,d) with these better comparisons and moved the original figure panels to the supplementary materials.

The following has been added in the text (Page 23 in the revised manuscript with track changes): "At two China coastal sites, Fuzhou and Hong Kong, the model results at the corresponding grid boxes are much lower than the observations (Fig. S4). We tried sampling the model results at adjacent gridboxes and found that those for the gridbox to the west are much more comparable to the observed (Fig. 9c and 9d). This suggests that the observations at both sites are significantly affected by local  $^{222}\text{Rn}$  emissions. The  $^{222}\text{Rn}$  observations show a minimum in summer, reflecting the intrusion of low- $^{222}\text{Rn}$  marine air associated with the Asian summer monsoon. Although the model successfully captures the observed seasonality, the simulation with ZKC (with enhanced emissions in China) shows a much better agreement compared to the large low bias in the simulation with JA97."

Revised Figure 9:

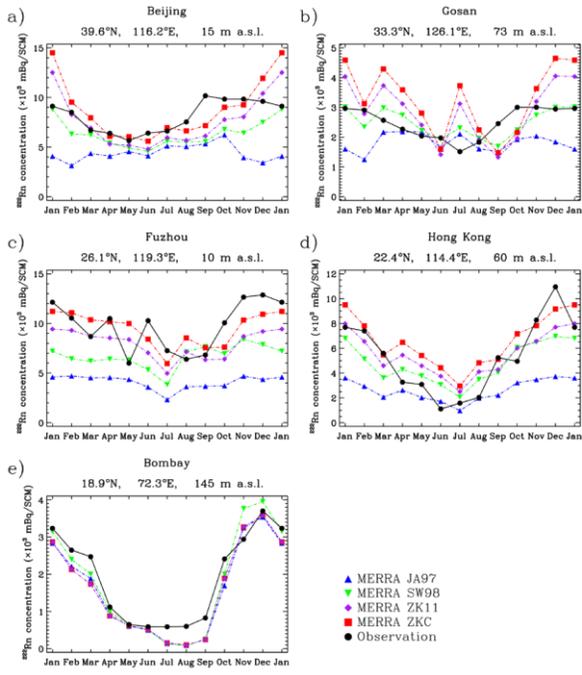


Figure 9. Same as Fig. 8, but for Asia. Note that the model results used in panel c) Fuzhou and d) Hong Kong are sampled at the gridboxes to the west of the ones where the sites are located to achieve a better agreement with the observations. See text for details.

In the supplementary materials, we added:

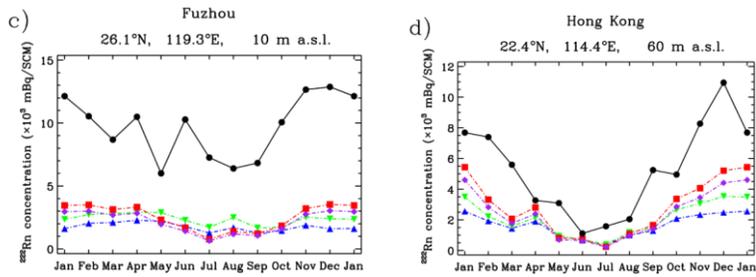


Figure S4. Same as Fig. 9 (c) and (d), but the simulated  $^{222}\text{Rn}$  concentrations are sampled at the model gridboxes corresponding to the site locations.

**Simulation of radon-222 with the GEOS-Chem global model:**

**Emissions, seasonality, and convective transport**

5 Bo Zhang<sup>1</sup>, Hongyu Liu<sup>1</sup>, James H. Crawford<sup>2</sup>, Gao Chen<sup>2</sup>, T. Duncan Fairlie<sup>2</sup>, Scott Chambers<sup>3</sup>,  
Chang-Hee Kang<sup>4</sup>, Alastair G. Williams<sup>3</sup>, Kai Zhang<sup>5</sup>, David B. Considine<sup>6</sup>, Melissa  
P. Sulprizio<sup>7</sup>, and Robert M. Yantosca<sup>7</sup>

<sup>1</sup>National Institute of Aerospace, Hampton, Virginia, USA

10 <sup>2</sup>NASA Langley Research Center, Hampton, Virginia, USA

<sup>3</sup>Australian Nuclear Science and Technology Organization, Kirrawee, New South Wales, Australia

<sup>4</sup>Jeju National University, Jeju, Republic of Korea

<sup>5</sup>Pacific Northwest National Laboratory, Richland, Washington, USA

<sup>6</sup>NASA Headquarters, Washington, D.C., USA

15 <sup>7</sup>Harvard University, Cambridge, Massachusetts, USA

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*Correspondence to:* Hongyu Liu (hongyu.liu-1@nasa.gov)

20 **Abstract.** Radon-222 (<sup>222</sup>Rn) is a short-lived radioactive gas naturally emitted from land surfaces, and has  
long been used to assess convective transport in atmospheric models. In this study, we simulate <sup>222</sup>Rn using  
the GEOS-Chem chemical transport model to improve our understanding of <sup>222</sup>Rn emissions and surface  
concentration seasonality, and characterize convective transport associated with two Goddard Earth  
Observing System (GEOS) meteorological products, MERRA and GEOS-FP. We evaluate four global  
25 <sup>222</sup>Rn emission scenarios by comparing model results with observations at 51 surface sites. The default

emission scenario in GEOS-Chem yields a moderate agreement with surface observations globally (68.9% of data within a factor of 2) and a large underestimate of winter surface  $^{222}\text{Rn}$  concentrations at Northern Hemisphere mid- and high-latitudes due to an oversimplified formulation of  $^{222}\text{Rn}$  emission fluxes ( $1 \text{ atom cm}^{-2} \text{ s}^{-1}$  over land with a reduction by a factor of 3 under freezing conditions). We compose a new global  $^{222}\text{Rn}$  emission scenario based on Zhang et al. (2011) and demonstrate its potential to improve simulated surface  $^{222}\text{Rn}$  concentrations and seasonality. The regional components of this scenario include spatially and temporally varying emission fluxes derived from previous measurements of soil radium content and soil exhalation models, which are key factors in determining  $^{222}\text{Rn}$  emission flux rates. However, large model underestimates of surface  $^{222}\text{Rn}$  concentrations still exist in Asia, suggesting unusually high regional  $^{222}\text{Rn}$  emissions. We therefore propose a conservative up-scaling factor of 1.2 for  $^{222}\text{Rn}$  emission fluxes in China, which was also constrained by observed deposition fluxes of  $^{210}\text{Pb}$  (a progeny of  $^{222}\text{Rn}$ ). With this modification, the model shows better agreement with observations in Europe and North America (>80% of data within a factor of 2), and reasonable agreement in Asia (close to 70%). Further constraints on  $^{222}\text{Rn}$  emissions would require additional concentration and emission flux observations in the central U.S., Canada, Africa, and Asia. We also compare and assess convective transport in model simulations driven by MERRA and GEOS-FP using observed  $^{222}\text{Rn}$  vertical profiles in northern mid-latitude summer, and from three short-term airborne campaigns. While simulations with both GEOS products are able to capture the observed vertical gradient of  $^{222}\text{Rn}$  concentrations in the lower troposphere (0-4 km), neither correctly represents the level of convective detrainment, resulting in biases in the middle and upper troposphere. Compared with GEOS-FP, MERRA leads to stronger convective transport of  $^{222}\text{Rn}$ , which is partially compensated for by its weaker large-scale vertical advection, resulting in similar global vertical distributions of  $^{222}\text{Rn}$  concentrations between the two simulations. This has important implications for using chemical transport models to interpret the transport of other trace species when these GEOS products are used as driving meteorology.

## 1 Introduction

A reasonable representation of transport processes in global models is critical to properly simulate tropospheric trace gases and aerosols. However, convective transport and boundary-layer turbulent mixing occur at sub-grid scales and are usually parameterized, unavoidably introducing transport biases. Radon-222 ( $^{222}\text{Rn}$ , half-life 3.8 days), an atmospheric radionuclide, is an excellent tracer for assessing these biases due to its relatively well-constrained sources and fairly simple sink pathway (radioactive decay) in the atmosphere (Jacob et al., 1997; Liu et al., 1984). In this work, we evaluate and improve the simulation of  $^{222}\text{Rn}$  in a global chemical transport model (GEOS-Chem CTM) and assess the role of convective transport in shaping [its  \$^{222}\text{Rn}\$](#)  vertical distributions.

Radon-222 is an inert gas ubiquitously produced in soils and rocks by radioactive decay of radium ( $^{226}\text{Ra}$ ). Shortly after  $^{222}\text{Rn}$  emanates to the atmosphere it decays to  $^{210}\text{Pb}$  (half-life 22.3 years). Wet and dry depositions of  $^{222}\text{Rn}$  are negligible due to its inert nature. The spatial distribution of  $^{222}\text{Rn}$  is therefore strongly affected by convective and synoptic-scale transport. Numerous studies have used  $^{222}\text{Rn}$  to evaluate model transport processes, such as boundary-layer structure and stability, vertical motion and mixing, and convection. For instance, Liu et al. (1984) derived seasonal vertical eddy diffusion coefficients using observed vertical profiles of tropospheric  $^{222}\text{Rn}$  concentrations. Allen et al. (1996) used  $^{222}\text{Rn}$  profile measurements to evaluate moist convection in their model and showed that deep convection from the boundary layer to the upper troposphere facilitates the formation of a “C-like”  $^{222}\text{Rn}$  vertical profile. Considine et al. (2005) used  $^{222}\text{Rn}$  and  $^{210}\text{Pb}$  measurements to examine the roles of convective transport in three different meteorological data sets. Zhang et al. (2008) tested two widely used convection schemes, Zhang-McFarlane-Hack (Hack, 1994; Zhang and McFarlane, 1995) and Tiedtke-Nordeng (Nordeng, 1994), in a global circulation model against observed  $^{222}\text{Rn}$  profiles. Although model results with both schemes showed similarly reasonable estimates of surface  $^{222}\text{Rn}$  concentrations, some [degree of](#) discrepancies were found in the middle and upper troposphere.  $^{222}\text{Rn}$  has also been used as an indicator of continental

influences on remote marine regions (Balkanski et al., 1992; Chambers et al., 2013, 2018). Model inter-comparison of simulated  $^{222}\text{Rn}$  distributions has been an efficient approach to compare and contrast transport characteristics with respect to boundary-layer turbulent mixing and convection (Genthon and Armengaud, 1995; Jacob et al., 1997).

5  $^{222}\text{Rn}$  emission fluxes have been estimated based on: (1) direct measurements, usually by assuming a linear increase of  $^{222}\text{Rn}$  in a chamber placed on soil, and (2) indirect estimates, through an integration of  $^{222}\text{Rn}$  profiles by assuming a local balance between  $^{222}\text{Rn}$  emission and decay. Using both approaches, Wilkening et al. (1972, 1975) derived an estimate of global mean  $^{222}\text{Rn}$  emission fluxes ( $0.75 \text{ atom cm}^{-2} \text{ s}^{-1}$  over land). Turekian et al. (1977) later suggested this global mean flux rate was likely an underestimate due  
10 to the assumption of a local steady state. By also considering one-dimensional longitudinal transport, Turekian et al. (1977) recommended a higher global mean flux of  $1.2 \text{ atom cm}^{-2} \text{ s}^{-1}$ , which led to a better agreement with observed  $^{210}\text{Pb}$  deposition fluxes across various latitudes. More recently, a mean global emission flux of  $1.0 \text{ atom cm}^{-2} \text{ s}^{-1}$  was considered more accurate, and has been used uniformly over land as a standard configuration (Balkanski et al., 1993).  $^{222}\text{Rn}$  fluxes from water surface are a few orders of  
15 magnitude lower and can be neglected compared with emissions over land (Wilkening and Clements, 1975). To date, most global models have used a globally uniform  $^{222}\text{Rn}$  emission flux of  $1.0 \text{ atom cm}^{-2} \text{ s}^{-1}$  with modifications in high latitudes and for freezing soil temperatures.

Quantification of regional and temporal  $^{222}\text{Rn}$  emission variations has been extended to broader areas and improved by new measurement techniques and modeling approaches. Observations have indicated that  
20 local  $^{222}\text{Rn}$  emission fluxes vary extensively with surface texture, soil moisture, radium content, ice coverage, and freezing condition (Martell, 1985; Turekian et al., 1977). The increasing availability of observational data inspired studies to quantify regional and temporal emission variations. Based on a large collection of global observations, Conen and Robertson (2002) proposed a linearly decreasing gradient in the Northern Hemisphere, from  $1 \text{ atom cm}^{-2} \text{ s}^{-1}$  at  $30^\circ\text{N}$  to  $0.2 \text{ atom cm}^{-2} \text{ s}^{-1}$  at  $70^\circ\text{N}$ . Regional and global  
25  $^{222}\text{Rn}$  emission flux datasets at fine resolution have also been developed based on models of gas diffusion

in porous media; this was facilitated by increasingly available soil parameters from meteorological models and assimilation (or reanalysis) datasets. Genthon and Armengaud (1995) introduced soil parameters into a global GCM to formulate online soil-atmosphere exchange of  $^{222}\text{Rn}$ , which assisted in capturing rapid fluctuations of surface  $^{222}\text{Rn}$  concentrations over freezing surfaces. Zhuo et al. (2008) compiled radium content information from over a thousand sites in China and constructed a high spatial resolution emission map over the country. Hirao et al. (2010) constructed a decade-long global  $^{222}\text{Rn}$  emission record based on additional considerations about surface texture. ~~Due to the availability of extensive measurements of  $^{222}\text{Rn}$  emission fluxes and surface concentrations, Europe has the Published  $^{222}\text{Rn}$  finest resolution~~ inventories for Europe y have very fine spatial resolutions (of up to  $0.083^\circ \times 0.083^\circ$ ) with monthly variability due to extensive measurements of emission fluxes and surface concentrations across the continent ~~(in regional and temporal emissions~~ (Karstens et al., 2015; López-Coto et al., 2013; Szegvary et al., 2009). Such variability is missing in the current GEOS-Chem standard model, which limits the use of  $^{222}\text{Rn}$  as a tracer to evaluate model transport processes, not to mention that  $^{222}\text{Rn}$  emission and distributions directly affect the production of its progeny  $^{210}\text{Pb}$ , a useful tracer for testing aerosol transport and wet deposition (Liu et al., 2001; Considine et al., 2005).

GEOS-Chem is driven by assimilated meteorological data sets archived from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office (GMAO). Changes in the model dynamics often occur as the GEOS model evolves, which in turn affect the characteristics of transport of chemical species in GEOS-Chem. In an evaluation with satellite observed carbon monoxide in the upper troposphere and lower stratosphere (UTLS), Liu et al. (2013) reported that less carbon monoxide was lofted to the UTLS when GEOS-Chem was driven by the GEOS-5 assimilated data due to insufficient vertical transport compared with GEOS-4. Downward transport from stratosphere to troposphere was previously found to be substantially overestimated in CTMs driven by GEOS-1 compared with GEOS-4 (Liu et al., 2016). In a similar manner, evaluation of  $^{222}\text{Rn}$  simulations with observations will help characterize convective transport and its uncertainty in the GEOS series.

In this paper, we assess and improve the simulation of  $^{222}\text{Rn}$  as a model utility to test convective transport in GEOS-Chem. We incorporate into the model recently published global  $^{222}\text{Rn}$  emission scenarios. We conduct model simulations with varying emission configurations and provide insights into potential biases in regional and seasonal emissions through evaluations against observed surface  $^{222}\text{Rn}$  concentrations and vertical profiles. We also present the ~~apparent~~ changes in simulated  $^{222}\text{Rn}$  vertical distributions as the driving meteorology switches between the Modern-Era Retrospective analysis for Research and Applications (MERRA) and GEOS Forward Processing (GEOS-FP), with a specific focus on the role of convection.

The rest of this paper is organized as follows. Section 2 describes the GEOS-Chem model, four  $^{222}\text{Rn}$  emission scenarios, model simulations, and observational data sets used in this work. Section 3 evaluates the four different  $^{222}\text{Rn}$  emission scenarios by comparing simulated  $^{222}\text{Rn}$  with surface measurements. Section 4 discusses potentially ~~excessive-underestimated~~  $^{222}\text{Rn}$  emissions in Asia. Section 5 examines simulated surface  $^{222}\text{Rn}$  seasonality at selected sites. Section 6 assesses convective transport in the model and compares the role of convective transport in MERRA and GEOS-FP in the  $^{222}\text{Rn}$  vertical distribution.

## 2 Model and data

### 2.1 GEOS-Chem

GEOS-Chem (<http://www.geos-chem.org>) is a global 3-D CTM of atmospheric composition with aerosol-chemistry interactions in both the troposphere and stratosphere, driven by GEOS assimilated meteorological fields from the NASA GMAO (e.g., Bey et al., 2001; Park et al., 2004; Eastham et al., 2014; ). The model uses ~~a flux-form semi-Lagrangian finite volume scheme, known as the~~ TPCORE, ~~advection algorithm of to calculate advection~~ (Lin and Rood, 1996) ~~Lin and Rood (1996)~~. The scheme uses ~~the monotonic piecewise parabolic method under convergence conditions and a semi-Lagrangian method otherwise~~. Convective transport is calculated using archived convective mass fluxes (Wu et al., 2007). Boundary-layer mixing is based on the non-local scheme implemented by Lin and McElroy (2010). In this

study, we use two different GEOS products (MERRA and GEOS-FP) to drive the model simulations. MERRA is a 30-year reanalysis product based on GEOS-5.2.0 (Rienecker et al., 2011). Its native resolution is  $0.667^\circ$  longitude by  $0.5^\circ$  latitude, with 72 vertical layers from the surface up to 80 km. GEOS-FP is the current operational product of GEOS-5.7 (and after) using an analysis developed jointly with NOAA's National Centers for Environmental Prediction (NCEP). It has a native resolution of  $0.3125^\circ$  longitude by  $0.25^\circ$  latitude with the same vertical grids as MERRA. Both the MERRA and GEOS-FP fields are regridded to the resolution of  $2.5^\circ$  longitude by  $2^\circ$  latitude (with vertical layers reduced to 47 levels) to drive In All GEOS-Chem simulations for in this study used a horizontal resolution of we used the MERRA and GEOS-FP fields regridded to  $2.5^\circ$  longitude by  $2^\circ$  latitude with vertical layers reduced to 47 levels, and the MERRA and GEOS-FP fields were regridded to the same resolution to drive the simulations. The meteorological archives have temporal resolutions of 3 h for 3-D fields and 1 h for 2-D fields. MERRA and GEOS-FP use similar model schemes for fundamental dynamical and physical processes. They both use the modified Relaxed Arakawa-Schubert scheme for convection (Moorthi and Suarez, 1992) and a combined turbulence parameterization based on Lock et al. (2000) and Louis et al. (1981). Compared with MERRA, GEOS-FP made a few adjustments including, but not limited to, increasing re-evaporation in precipitation and adjusting the balance between local and non-local turbulent diffusion, with the former resulting in a considerable increase in water vapor in the tropical troposphere (Molod et al., 2012). MERRA-2, which is based on a newer version of GEOS-5 and shows improved climate over MERRA (Molod et al., 2015), is not used here since it was not ready to drive our model when this study was started.

GEOS-Chem (v11-01f, [http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-Chem\\_v11-01](http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-Chem_v11-01)) includes a radionuclide ( $^{222}\text{Rn}$ - $^{210}\text{Pb}$ - $^7\text{Be}$ ) simulation option, which runs independently from the full oxidant-aerosol chemistry simulation. The radionuclide tracers have been used to evaluate chemical transport (Jacob et al., 1997; Yu et al., 2018) and wet deposition processes (Liu et al., 2001, 2004) in the

model. The simulation of  $^{222}\text{Rn}$  includes emissions, transport (advection, convection, boundary layer mixing), and radioactive decay. Wet and dry deposition of  $^{222}\text{Rn}$  are neglected in the model.

## 2.2 $^{222}\text{Rn}$ emission scenarios

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The standard version of GEOS-Chem uses the  $^{222}\text{Rn}$  emission scenario of Jacob et al. (1997) (hereafter referred to as JA97). The World Climate Research Program (WCRP) Cambridge Workshop of 1995 (Rasch et al., 2000) previously used JA97 to compare  $^{210}\text{Pb}$  deposition processes in multiple atmospheric models. JA97 was developed using the estimated global mean  $^{222}\text{Rn}$  fluxes of Turekian et al. (1977) and only considered emission variations for a few broad latitude bands. The  $^{222}\text{Rn}$  emission fluxes in JA97 are uniformly set to be  $1 \text{ atom cm}^{-2} \text{ s}^{-1}$  over land between  $60^\circ\text{N} - 60^\circ\text{S}$ ,  $0.005 \text{ atom cm}^{-2} \text{ s}^{-1}$  between  $60^\circ\text{N} - 70^\circ\text{N}$  and  $60^\circ\text{S} - 70^\circ\text{S}$ , and zero poleward of  $70^\circ\text{N/S}$ . Emission fluxes over lakes and oceans are set to  $0.005 \text{ atom cm}^{-2} \text{ s}^{-1}$ . Emissions are reduced by a factor of 3 when surface temperature is below  $0^\circ\text{C}$  on account of the depressed exhalation of  $^{222}\text{Rn}$  under freezing conditions. Such a temperature-dependent reduction can lead to underestimated emissions in winter because soils may not be totally frozen when temperature falls below  $0^\circ\text{C}$  for only a short period of time. The overall uncertainty of the JA97 emission was estimated to be within 25% globally (Jacob et al., 1997). [Since the emission fluxes in JA97 are fairly uniform over land area, this simplistic emission scenario has been can be used to measurediscern continental influence on air masses in global models and assess the effect of any changes in the model representation of convectionave mixingchemes in global models \(e.g., Balkanski et al., 1992; Jacob et al., 1997\).](#)

A few  $^{222}\text{Rn}$  emission scenarios were published after Jacob et al. (1997). Conen and Robertson (2002) proposed a  $^{222}\text{Rn}$  emission scenario having a uniform  $^{222}\text{Rn}$  flux of  $1 \text{ atom cm}^{-2} \text{ s}^{-1}$  from the continental surface in the Southern Hemisphere and tropics and a linear decreasing trend from  $1 \text{ atom cm}^{-2} \text{ s}^{-1}$  at  $30^\circ\text{N}$  to  $0.2 \text{ atom cm}^{-2} \text{ s}^{-1}$  at  $70^\circ\text{N}$ . This decreasing trend towards high latitudes was supported by experimental results showing a decrease of  $^{222}\text{Rn}$  fluxes in the local soil with higher water table and organic portion. This proposed latitudinal gradient was found to be in a good agreement with an estimated value based on multi-

year observations at a few Asian sites (Williams et al., 2009). However, this emission scenario is not used in this work because it does not include regional variations other than the linear latitudinal gradient.

Schery and Wasiolek (1998, hereafter SW98) published the first global  $^{222}\text{Rn}$  emission inventory that included detailed regional and seasonal variations on a monthly basis (at  $1^\circ$  longitude by  $1^\circ$  latitude resolution). The emission flux in SW98 is formulated by using a theoretical diffusion model of porous soil with controlling factors of soil radium content, soil moisture, and surface temperature. The estimated annual global average  $^{222}\text{Rn}$  emission flux was  $1.63 \pm 0.43 \text{ atom cm}^{-2} \text{ s}^{-1}$ , higher than the widely used JA97 constant value of  $1 \text{ atom cm}^{-2} \text{ s}^{-1}$ . Global  $^{222}\text{Rn}$  emissions in SW98 exhibited regional variations of a factor of 3 and seasonal variations of a factor of 2. The dominant factor in determining the regional variations in  $^{222}\text{Rn}$  fluxes was found to be the soil radium concentrations according to Schery and Wasiolek (1998). Emission fluxes in the U.S. and China feature more detailed regional variations because soil radium concentrations in these countries were incorporated. However, it was suggested that SW98 overestimated the global average  $^{222}\text{Rn}$  flux (Koch et al., 2006; Zhang et al., 2011). When using the SW98 emissions in a global model simulation of  $^{210}\text{Pb}$ , Koch et al. (2006) found it necessary to reduce the emissions by half to improve the excessive  $^{210}\text{Pb}$  concentrations in their model. In this study, we reduce the emission fluxes of SW98 globally by a factor of 1.6, as recommended by Zhang et al. (2011).

Zhang et al. (2011, hereafter ZK11) compiled a new global  $^{222}\text{Rn}$  emission inventory based on a combination of SW98 (with a global reduction factor of 1.6) and recently published  $^{222}\text{Rn}$  flux measurements in Europe and the U.S. (Szegvary et al., 2007), China (Zhuo et al., 2008), Australia (Griffiths et al., 2010), and -oceanic regions (Schery and Huang, 2004). In ZK11,  $^{222}\text{Rn}$  emissions in Europe were derived from a demonstrated linear relationship between terrestrial gamma dose rate and  $^{222}\text{Rn}$  emissions (Szegvary et al., 2007). The relationship allows a convenient calculation of regional  $^{222}\text{Rn}$  emissions for places where automatic measurements of gamma dose rate have been established, e.g., in Europe (Szegvary et al., 2009). This  $^{222}\text{Rn}$  emission inventory for Europe has recently been updated (Karstens et al., 2015; López-Coto et al., 2013) with further detailed information on soil and surface

roughness and minor modifications about handling  $^{222}\text{Rn}$  transport in porous media. A high-resolution (25 km  $\times$  25 km)  $^{222}\text{Rn}$  emission map for China was included in ZK11 based on work by Zhuo et al. (2008), who estimated the nation-wide emissions according to measurements of radium content in surface soil at 1099 sites in China. The oceanic emission flux used was  $0.00182 \text{ atom cm}^{-2} \text{ s}^{-1}$ , derived by Schery and Huang (2004) with a gas transfer model, significantly lower than typical  $^{222}\text{Rn}$  emissions over land. ~~In For each model surface grid-boxes containing both land and water surfaces, we summed  $^{222}\text{Rn}$  emission fluxes from both land and water surface types according to weighted by the their respective land and water areas of land or water.~~

In this study, we modify ZK11 to a customized  $^{222}\text{Rn}$  emission scenario (hereafter referred to as ZKC) and constrain the inventory with observations of surface  $^{222}\text{Rn}$  concentrations. This customized emission scenario adopts ZK11 for most areas except for North America, where the SW98 emission fluxes are used with a reduction factor of 1.6, following previous model studies (Koch et al., 2006; Zhang et al., 2011). We also increase the emission over China by a factor of 1.2 all year round due to potentially ~~excessive underestimated~~  $^{222}\text{Rn}$  emission there, which will be discussed in detail in Section 3.2. The emission enhancement factor is only tentative due to very few surface  $^{222}\text{Rn}$  measurements available in western China and a lack of seasonality in the measurements. ~~Since ZK11 has been tested and lead to a with satisfactory agreements between with modeled and observed surface concentrations in Europe (Zhang et al., 2011). The the updates for emission fluxes in Europe by López-Coto et al. (2013) and Karstens et al. (2015) are not included. The largest terrestrial spatial variation of  $^{222}\text{Rn}$  emission rates in ZKC is a factor of~~ 10. ~~The descriptions for of the four emission scenarios are summarized is given in Table 1.~~

Figure 1 shows the global annual mean  $^{222}\text{Rn}$  emission fluxes of the four emission scenarios described above. Compared with the standard GEOS-Chem  $^{222}\text{Rn}$  emission scenario (JA97, Fig. 1a), the other three show evident spatial variations of varying extents due to incorporation of observations and estimates from soil exhalation models. The estimated global total  $^{222}\text{Rn}$  emissions for JA97, SW98, ZK11, and ZKC are  $1.94 \text{ GCi (Giga-Curie) year}^{-1}$ ,  $2.41 \text{ GCi year}^{-1}$ ,  $2.11 \text{ GCi year}^{-1}$ , and  $2.22 \text{ GCi year}^{-1}$ , respectively. Since

there is no consensus on the global total  $^{222}\text{Rn}$  emission, we do not normalize the total emission amount for each scenario. Instead, the overall evaluation of the emission scenarios is based on comparisons with surface  $^{222}\text{Rn}$  observations. It is clearly shown in Fig. 1 that the three later  $^{222}\text{Rn}$  emission scenarios have substantial enhancements of  $^{222}\text{Rn}$  emission fluxes in North America and East Asia. SW98 exhibits more intense  $^{222}\text{Rn}$  emissions in North America, which have been adopted in ZKC. In the northern polar region, SW98 presents much higher  $^{222}\text{Rn}$  emissions over Siberia extending to higher latitudes. JA97 is overly simplified and has nearly no emissions over Siberia due to temperature-dependent reduction in the cold high-latitude regions. The ZK11 emissions in Siberia stay between those of JA97 and SW98, with somewhat higher emissions in the eastern Siberia. ZK11 has much higher  $^{222}\text{Rn}$  emissions in China, which are further scaled up by a factor of 1.2 in ZKC.

Seasonal variations of  $^{222}\text{Rn}$  emissions are considered in all four scenarios but with different approaches. In JA97,  $^{222}\text{Rn}$  emissions are reduced by a factor of 3 when surface temperature in the driving meteorological fields falls below  $0\text{ }^{\circ}\text{C}$ , thus resulting in seasonal variations of  $^{222}\text{Rn}$  emissions in high-latitude regions. In the other emission scenarios, the monthly varying  $^{222}\text{Rn}$  emission fluxes in each model bottom layer are prescribed based on observed and assumed soil parameters (see SW98) and do not change from year to year. Figures 2 and 3 compare monthly mean  $^{222}\text{Rn}$  emissions for January and July in the emission inventories.  $^{222}\text{Rn}$  emissions are generally the lowest in January because of the inhibition of exhalation as a result of ice cover and high moisture content. All emission scenarios exhibit increased global  $^{222}\text{Rn}$  emissions by a factor of 1.2 to 1.4 in July compared with January due to enhanced emissions over the Northern Hemisphere continents. The summer-winter changes of local emissions are mostly within a factor of 2. The possible underestimation of emissions for surface temperatures under  $0\text{ }^{\circ}\text{C}$  is revised in the later emission scenarios, leading to increased wintertime emissions in central and eastern Asia, North America, and southern Europe (Fig. 2a vs 2d). The affected regions extend to ~~further~~ lower latitudes in eastern Asia and North America compared with relatively warmer Europe.

Compared to JA97, significant emission increases occur in mid-low latitudes where land is covered by desert or mountainous texture in the later emission scenarios, e.g., western U.S. and western China. Rocky and desert land types are more favorable for  $^{222}\text{Rn}$  emission compared with soil. Previous related literature and analyses support these emission modifications based on evaluations against existing surface observations. We speculate that some degree of emission increase would be reasonable in the Middle East and North Africa where land is mostly covered by desert. Emissions in these areas in ZK11 and ZKC are adapted from SW98, which uses a world average surface radium content to calculate  $^{222}\text{Rn}$  emission. No observations of surface radium content or  $^{222}\text{Rn}$  concentrations exist for evaluating speculated emission modifications, but future changes are possible [when measurements become available depending on the availability of measurements](#) in these areas.

### 2.3 Model simulations and observational data

We simulated  $^{222}\text{Rn}$  with the model driven by MERRA using the four emission scenarios. The preferred emission scenario was then identified based on a comparison of simulated and observed surface  $^{222}\text{Rn}$  concentrations and seasonality. To characterize convective transport in the MERRA and GEOS-FP products, we also conducted model sensitivity experiments for which convective transport was turned off. All simulations were conducted for the year of 2013 with a 12-month spin-up, which was initialized with a climatological restart file from previous model simulations. Table [24](#) lists all the model experiments and their configurations.

We evaluate the 2013 simulations against long-term monthly or annual  $^{222}\text{Rn}$  observations. We used the observed surface  $^{222}\text{Rn}$  concentration dataset compiled by Zhang et al. (2011), who evaluated the ZK11 emission scenario in their model. Figure 4 shows the locations of 51 surface  $^{222}\text{Rn}$  measurement sites. The sites are concentrated in Europe, North America, and East Asia. Fewer sites [\(11\)](#) are located in the Southern Hemisphere. No observations in boreal Canada and Siberia are available. The few in-land sites in China only reported annual means [based on measurements of 1-2 years \(Jin et al., 1998\)](#). The  $^{222}\text{Rn}$  observations were made in [consecutive-multiple](#) years, and we treat the calculated multi-year monthly

means as climatological. We also include longer period observations at Mauna Loa (2004-2010; Chambers et al., 2016c) and Gosan (2001-2010; Chambers et al. 2016b) stations in addition to those compiled by Zhang et al. (2011) in our analyses below. Considering the monthly climatological surface  $^{222}\text{Rn}$  observations used in the comparisons, simulations driven by MERRA for alternative years do not change the conclusions of this study.

To examine simulated convective transport characteristics, we compare model results with four observational datasets of  $^{222}\text{Rn}$  vertical profiles (Liu et al., 1984; Zaucker et al., 1996; Kritz et al., 1998; Williams et al., 2011). The scarcity of  $^{222}\text{Rn}$  airborne measurements is partly due to the fact that the measurement requires an extraction and counting facility nearby in order to minimize decay and that the process of radon extraction is labor-intensive (Williams et al., 2011). Liu et al. (1984) compiled an extensive dataset of  $^{222}\text{Rn}$  profiles for different seasons based on airborne measurements made in the 1950s-1970s. The summertime average profile was calculated from 23 sites in the U.S., Ukraine, and central Asia, and mainly represents the summertime  $^{222}\text{Rn}$  vertical distribution over the Northern Hemisphere mid-latitude continental regions. Zaucker et al. (1996) reported nine  $^{222}\text{Rn}$  profiles measured during flights from east coast of Canada (Nova Scotia and New Brunswick) to the North Atlantic as part of the North Atlantic Regional Experiment (NARE, August 1993). Kritz et al. (1998) measured  $^{222}\text{Rn}$  vertical profiles at Moffett Field (37.4 °N, 122.0 °W), a coastal site in California, U.S., during April to August in 1994. The Moffett profile represents summertime  $^{222}\text{Rn}$  vertical distribution over an offshore region. Williams et al. (2011) made aircraft measurements of  $^{222}\text{Rn}$  profiles up to 3.5 km altitude at Goulburn (34.8°S, 149.7°E), an inland rural site in New South Wales, Australia, during May 2006-2008 and January 2007 (Williams et al., 2011).

### 3 Model results and evaluation with surface observations

#### 3.1 Model surface $^{222}\text{Rn}$

Figure 5a-b show the global surface  $^{222}\text{Rn}$  concentrations for January and July 2013, as simulated by GEOS-Chem with the JA97 emission scenario (simulation A1, Table 24). Surface  $^{222}\text{Rn}$  concentrations are much higher over land at low- and mid-latitudes compared with marine areas and high-latitudes. Typical surface  $^{222}\text{Rn}$  concentrations over land range from a few hundreds to about  $1.0 \times 10^4 \text{ mBq SCM}^{-1}$ . Surface concentrations drop sharply from land to oceanic regions due to the short lifetime of  $^{222}\text{Rn}$ , with the values over the oceans ~2-3 magnitudes lower and ranging from tens to a few hundreds of  $\text{mBq SCM}^{-1}$ . The model simulates a noticeable outflow of  $^{222}\text{Rn}$  at surface level from the west coast of Africa to South America in January. Surface  $^{222}\text{Rn}$  concentrations are higher overall in winter due to shallower boundary layers than in summer (Fig. 5a vs Fig. 5b). For example, concentrations in Europe, central and East Asia, and North Africa are ~~significantly lower~~ by a few thousands  $\text{mBq SCM}^{-1}$  in July (summer), while concentrations in South Africa, Argentina and Australia are ~~substantially higher~~ by similar magnitudes in July (winter). The contrasting seasonality of surface  $^{222}\text{Rn}$  concentrations (high in winter) compared to emission fluxes (high in summer) suggests that the accumulation effect in shallower boundary layers (weakened vertical transport and mixing, see Fig. S1) dominates the seasonal changes in emission when it comes to determining the seasonality of surface  $^{222}\text{Rn}$  concentrations.

Figure 5c-h show the changes in simulated surface  $^{222}\text{Rn}$  concentrations when the SW98, ZK11, and ZKC emissions are used (simulation A2-A4, Table 24), relative to the standard simulation with the JA97 emissions. All three alternative  $^{222}\text{Rn}$  emission scenarios lead to remarkable increases in surface concentrations in mid- and high-latitude regions of North America and Asia. With SW98 (simulation A2, Fig. 5c), ~~significantly increased~~ winter surface  $^{222}\text{Rn}$  concentrations increased from very low levels ( $<1000 \text{ mBq SCM}^{-1}$ ) to about  $1.0 \times 10^4 \text{ mBq SCM}^{-1}$  in many high-latitude regions, including northwestern U.S., Alaska, northern Canada, as well as in the continental areas extending from eastern Europe through Siberia to the Bering Strait. These large increases are mainly due to the zero emission flux

rate prescribed for high latitudes ( $> 60^\circ\text{N}$ ) in JA97, which is replaced in SW98 by fluxes from 0.3 to 0.6  $\text{atom cm}^{-2} \text{s}^{-1}$  (Fig. 2b and 3b). As shown later, this characteristic in JA97 overly simplifies  $^{222}\text{Rn}$  emission variations and causes underestimation of surface  $^{222}\text{Rn}$  concentrations in high-latitude regions in winter. Accumulation of  $^{222}\text{Rn}$  in the shallow winter boundary layer also contributes to and enhances the differences in surface  $^{222}\text{Rn}$  concentration caused by increased emissions. In the ZK11 simulation, similar enhancements of surface  $^{222}\text{Rn}$  appear in North America, China, and the far East Siberia (Fig. 5e, f), but the overall magnitudes of enhancement are smaller than those with SW98. The largest enhancements in Asia shift to the east and are seen in eastern Siberia rather than the whole boreal Siberia. ZK11 incorporates recent  $^{222}\text{Rn}$  flux measurements in Asia (Zhang et al., 2008) and shows some smaller changes from those of JA97 in Siberia. Since ZKC and SW98 share the same emissions for North America, the surface concentration changes are mostly identical between the two. For the same reason, the ZKC and ZK11 results look similar in Asia, except that the surface concentrations over China are more enhanced due to the upscaling in ZKC. In July (Fig 5d, f, h), the changes in surface  $^{222}\text{Rn}$  concentrations are much less significant for all emission scenarios. This also reflects the strong effects of summer boundary layer ventilation, which largely compensates for the differences caused by the seasonal emission changes.

### 3.2 Evaluation of emission scenarios with surface observations

Following Zhang et al. (2011), we evaluate the  $^{222}\text{Rn}$  emission scenarios by comparing model results with surface observations of  $^{222}\text{Rn}$  concentrations. We conduct the comparisons in the form of scatter plots for Europe (EU), Asia (AS), North America (NA), and the global (ALL), respectively (Fig. 6). For each observed monthly or annual mean, model output was sampled in the grid cell corresponding to the physical location and elevation of each site and then averaged for the corresponding observation time period. Also shown in Fig. 6 are the reduced-major-axis linear correlation coefficients (R; Hirsch and Gilroy (1984)) and the percentages (P) of the data points lying within a range of a factor of 2 (dashed lines).

Europe is the continent where emission fluxes and transport of  $^{222}\text{Rn}$  have been studied most extensively. The measurements are more widely and evenly distributed across the continent (Fig. 4). The

JA97 simulation (A1, Table 2+) shows moderate agreement with observations (P = 66.5%) bearing some large underestimates (Fig. 6a). The SW98 simulation has the lowest P value of 61.9% (Fig. 6b) due to a large number of points with high biases. ZK11 and ZKC use the same <sup>222</sup>Rn emissions in Europe, and the P values are close (80.3% and 80.7% in Fig. 6c and Fig. 6d, respectively). The better agreement when using ZK11 and ZKC substantiates the high resolution <sup>222</sup>Rn emission estimates derived from gamma dose rates in Europe (Szegvary et al., 2009). Schmithüsen et al. (2017) compared measured <sup>222</sup>Rn concentrations across the European sites in terms of different instruments and measurement systems and provided suggested scaling factors for each site. Here, the same evaluation for the emission scenarios with the scaling factors is given in Fig. S2. There are only slight changes in the P values for all regional groups, and the same rank of the four emission scenarios remains.

All simulations exhibit some degree of underestimation in Asia (Fig. 6e-h). Monthly mean observations are available for 7 of the 12 Asian sites, otherwise only annual means are available. Consequently, data points are sparse in Fig. 6e-h. The JA97 simulation shows poor agreement for Asia (P = 46.3%, Fig. 6e). Agreement for the others is better, but still deficient, with P values of 64.2%, 67.2%, and 68.7% for SW98, ZK11, and ZKC, respectively (Fig. 6f-h). The few underestimated data points in Fig. 6g and 6h are observed annual means from the inland Chinese sites. The observations in China were taken between 1 m and 1.5-m above ground according to Jin et al. (1998). The model surface layer concentrations usually represent the averages in the first-model bottom layer (~100 m high), and thus may be literally lower than the observations due to the steep concentration gradients near the surface, especially during nighttime (Chambers et al., 2011). On the other hand, there are possible low biases in the measurements; <sup>222</sup>Rn concentrations derived from <sup>222</sup>Rn progeny measurements (Schmithüsen et al., 2017; Grossi et al., 2020), compensating the high values taken near surface lessening the above model underestimate due to large near-surface vertical gradients. These biases differ on a case-by-case basis and are difficult to quantify. With upscaled emission in ZKC, the improvement compared with ZK11 is minor.

To better match the Asian observations, we tentatively conducted additional model simulations in which

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the Asian  $^{222}\text{Rn}$  emission fluxes are scaled up by a factor of 1.5 or 1.7 (instead of 1.2 in the recommended ZKC). The P values from those simulations are larger with some previously underestimated data moving into the 2-factor range; up-scaling by a factor of 1.5 would increase the P value to above 70%, but the simulated total  $^{210}\text{Pb}$  deposition fluxes at mid-latitudes would be substantially overestimated (Zhang et al.,

5 Constraints From Airborne Lead-210 Observations on Aerosol Scavenging and Lifetime in a Global Chemical Transport Model, manuscript in prep., 2020). As will be discussed in Section 4, a few studies reported unusually high surface concentrations and ~~excessive-large~~ emission fluxes at individual sites in Asia; the evidence therein endorsed a higher upscaling factor, which would reduce the model underestimates of surface concentrations. However, without knowing the distributions and varying extents

10 of emission biases in Asia, applying a higher and uniform scaling factor to the whole region may worsen the global simulation of  $^{210}\text{Pb}$  deposition. The few annual means that lead to the low P values may not be as representative as the monthly data and can be biased. Therefore, we use a tentative scaling factor of 1.2 for emission fluxes in China (i.e., ZKC) and expect future improvements when more observations of  $^{222}\text{Rn}$  emission and surface concentrations become available.

15 All simulations reproduced the observed surface  $^{222}\text{Rn}$  concentrations in North America well (Fig. 6i-l). SW98 (Fig. 6j) and ZKC (Fig. 6l) share identical  $^{222}\text{Rn}$  emissions over North America, and simulations with both emission scenarios show excellent agreement with the observations (P ~ 90%, Fig. 6j, l). This suggests that SW98 is an adequate option for  $^{222}\text{Rn}$  emissions in North America. Interestingly, ZKC leads to slightly better agreement compared with SW98, although identical emissions were used for North

20 America. A few overestimated data points in the simulation with SW98 are better simulated with ZKC at the U.S. west coast sites, as a result of the large reduction in emissions over the upwind Siberia regions (Fig. 1d). Despite the good agreement between model results and observations, the evaluation is limited to the western and eastern coastal regions of the U.S. Data from Africa, the central U.S., and Canada is currently lacking, and would otherwise improve the model evaluation, especially over North America.

Figures 6m-p show the overall evaluation of the model results against measurements at all 51 surface sites over the globe. Both ZK11 and ZKC simulations show better agreement with observations ( $P = 76.9\%$  and  $78.4\%$ , Fig. 6o, p), suggesting that ZK11 and ZKC are potentially better choices for replacing the JA97 emission scenario in the standard version of GEOS-Chem. Although with its ~~tentative-provisional~~ effort to address high Asian emissions ZKC is a step ahead of ZK11. The large biases of a few points outside the factor-of-2 range are from the Antarctic sites. None of the simulations can reasonably represent observations in Antarctica, which can be attributed to not well characterized emission (Chambers et al., 2018) and will be discussed later. If the two Antarctic sites (with model low biases in the lower left corner of Fig. 6o, p) were excluded, the  $P$  values for ZK11 and ZKC would increase to over  $80\%$ .

#### 4 Excessive $^{222}\text{Rn}$ emissions in East Asia

Unusually high  $^{222}\text{Rn}$  emissions have been observed over mainland Asia (Iida et al., 2000; Yamanishi et al., 1991) and downwind regions (e.g., Korea; Zahorowski et al., 2005). These high  $^{222}\text{Rn}$  emissions, not well represented in JA97-like emission scenarios, were likely responsible for the failure of CTMs in capturing the  $^{222}\text{Rn}$  concentrations observed in East Asia (Jacob et al., 1997). In particular,  $^{222}\text{Rn}$  emissions over China have been underestimated at inland sites (Zhang et al., 2011). China and India have been identified as regions of high  $^{222}\text{Rn}$  emissions from soil. It was suspected that this is partially due to high soil content of radium (Schery, 2004; Zhuo et al., 2008). Schery (2004) presented global measurements of radium content in soil, which clearly indicated that the radium concentrations are higher by about a factor of 3 in the southeastern compared to the northwestern China. Consistently, Zahorowski et al. (2005) found that surface  $^{222}\text{Rn}$  concentrations were roughly three times higher at Hok Tsui (Hong Kong) during winter compared to Gosan, where fetch is from northern China and Mongolia. Zhuo et al. (2008) provided an estimated area-weighted annual average  $^{222}\text{Rn}$  emission of  $29.7 \text{ mBq m}^{-2} \text{ s}^{-1}$  ( $\sim 1.41 \text{ atom m}^{-2} \text{ s}^{-1}$ ) in China. Based on three-year winter-time  $^{222}\text{Rn}$  observations at Sado Island, Japan, and associated trajectory analyses, Williams et al. (2009) suggested that emission fluxes can be 1.75 times higher in the lower

latitude bands over the Asian continent compared to higher latitudes. In an inverse modeling of Asian  $^{222}\text{Rn}$  emissions, Hirao et al. (2010) showed an area-weighted average  $^{222}\text{Rn}$  emission of  $33.0 \text{ mBq m}^{-2} \text{ s}^{-1}$  ( $\sim 1.57 \text{ atom m}^{-2} \text{ s}^{-1}$ ) in Asia with the highest emissions found in central and southeastern Asia. These values are considered much higher than typical  $^{222}\text{Rn}$  emission known for Europe, where Szegvary et al. (2009) suggested half of the continent has emissions ranging from  $8.33$  to  $14.6 \text{ mBq m}^{-2} \text{ s}^{-1}$  ( $0.40$  to  $0.70 \text{ atom m}^{-2} \text{ s}^{-1}$ ). Hirao et al. (2010) also found that, to better match surface observations at Hachijo Island, a volcanic island about 287 kilometers south of Tokyo in the Philippine Sea, the emissions over East Asia would need to be increased by a factor of 1.69.

It is likely that the high  $^{222}\text{Rn}$  emissions in Asia are poorly estimated because of the diverse climate and geographic textures formed on the largest continent of the earth. The southern part of China is known to be covered with soils containing higher radium concentrations than global average (Schery and Wasiolek, 1998). Central Asia is dry and sparsely covered with soils, which could facilitate  $^{222}\text{Rn}$  emanation. The mountainous surface in southeastern China could also be conducive to high  $^{222}\text{Rn}$  emissions. The  $^{222}\text{Rn}$  exhalation model developed by Hirao et al. (2010) took into consideration  $^{222}\text{Rn}$  emission enhancements caused by rough surfaces, but still underestimated  $^{222}\text{Rn}$  concentrations in East Asia. Active crust movements along the east coast of Asia can cause more exposure of radium and extra  $^{222}\text{Rn}$  emissions. Intense human activities may also contribute to excessive  $^{222}\text{Rn}$  emissions in Asia. Moore et al. (1976) pointed out that phosphate ores contain high concentration of  $^{238}\text{U}$  (precursor of radium) and are widely used as phosphate fertilizers in the populous East Asia region. Due to such complexities and uncertainties, most of the  $^{222}\text{Rn}$  exhalation models are not well validated in Asia, and a lack of  $^{222}\text{Rn}$  measurements in central and western Asia adds to the difficulty.

An alternative way to verify  $^{222}\text{Rn}$  emissions is to evaluate the deposition fluxes of its long-lived decay daughter,  $^{210}\text{Pb}$ . Since surface deposition is the primary sink of  $^{210}\text{Pb}$  aerosols, global  $^{210}\text{Pb}$  deposition fluxes should be balanced by  $^{210}\text{Pb}$  production or  $^{222}\text{Rn}$  emission fluxes (Considine et al., 2005). Regional total  $^{210}\text{Pb}$  deposition fluxes, however, can be affected by transport into and out of the region. Nevertheless,

comparisons between simulated and observed  $^{210}\text{Pb}$  deposition fluxes at multiple locations in Asia offer a test of ~~excessive-underestimated~~  $^{222}\text{Rn}$  emissions. Figure 7a compares model results with observed  $^{210}\text{Pb}$  total (dry and wet) deposition in Shanghai for each season averaged over an 8-year period (Du et al., 2015). All model simulations, including the simulation with upscaled emission in China (ZKC), underestimate the total deposition in all seasons. Enhanced  $^{222}\text{Rn}$  emissions in ZKC improve the simulated  $^{210}\text{Pb}$  deposition to a limited extent in all seasons and more favorably in winter. We then calculate the correlations between simulated and observed annual mean  $^{210}\text{Pb}$  deposition fluxes at the sites in North America (nine sites) and Asia (nine sites; Du et al. (2015)). Some studied Asian sites are located in Northern and inland China. Details about these sites can be found in Du et al. (2015). The reduced-major-axis regression slopes for North American sites are closer to 1 (Fig. 7b), indicating a generally well simulated lifecycle from  $^{222}\text{Rn}$  emission to  $^{210}\text{Pb}$  deposition. By contrast, the slopes for Asian sites are much lower than 1. This large magnitude of model underestimation in  $^{210}\text{Pb}$  deposition fluxes can only be attributed to low  $^{222}\text{Rn}$  emissions in Asia. Much existing evidence suggests using a larger scaling factor, but as mentioned earlier, we choose to use a moderate scaling factor of only 1.2 for China to avoid large overestimates of total  $^{210}\text{Pb}$  deposition fluxes ~~in~~ over the rest of the Northern Hemisphere.

### 5 Seasonality in surface $^{222}\text{Rn}$ concentrations

The seasonality in surface  $^{222}\text{Rn}$  concentrations is mainly affected by three factors: (1) the surface variability in  $^{222}\text{Rn}$  emission flux rate ~~determined by radium content and seasonally varying soil conditions~~ due to seasonal changes in soil moisture, diffusivity, depth of the water table, and snow and ice coverage; (2) the vertical mixing processes (i.e., boundary-layer mixing and convection); and (3) advective transport of  $^{222}\text{Rn}$ -rich or -poor air masses. The roles of these factors may vary by location. Here, we examine the seasonal variations of surface  $^{222}\text{Rn}$  concentrations at selected surface sites in Europe, Asia, and North America, and discuss these impacting factors. The selection of surface sites is mainly based on

the availability of multiple-year measurements, with consideration of special geographic locations indicative of regional transport patterns.

**Europe.** Observations in Europe were mostly obtained in Finland, Germany, France, and Italy, with about half of the sites in Finland. Figures 8a-c show the comparisons of model results with monthly mean observations at three Finland sites (Kevo, Pallas, and Joensuu). At these high-latitude sites, the highest monthly concentration does not exceed  $4000 \text{ mBq SCM}^{-1}$ , but the seasonal variations are large, with the observed wintertime highs being up to twice the summertime lows. Such seasonal variation is mainly due to shallower boundary layer and less convection in winter because the changes in  $^{222}\text{Rn}$  emissions are minor due to low temperature all year round (see Fig. 2 and 3). Szegvary et al. (2009) suggested that the  $^{222}\text{Rn}$  emissions in northern Europe are generally lower than the commonly used value of  $1 \text{ atom cm}^{-2} \text{ s}^{-1}$ . The soil water content is high because of the long snowy winter and short summer there. The content of radium is also found to be lower than average in the quaternary sand deposits. The ZK11 and ZKC emission scenarios, which adopted  $^{222}\text{Rn}$  emission fluxes derived from measured gamma radiation (Szegvary et al., 2009), are clearly the better options and result in better simulated seasonal variations (frequently overlapped purple and red lines in Fig. 8a-c). The SW98 emissions lead to much higher  $^{222}\text{Rn}$  concentrations compared with the observations, whereas JA97 tends to underestimate the emissions and results in lower concentrations.

Figure 8d-f show model-observation comparisons at three sites in central mainland Europe, i.e., Hohenpeissenberg (Germany), Freiburg (Germany), and Gif-sur-Yvette (France). The observations generally show minimal surface concentrations in spring and maximums in late fall. The highs appear earlier with larger seasonal amplitudes compared with the Finland sites as a result of combined effects of seasonal changes in emission fluxes and vertical transport. In general, the lowest  $^{222}\text{Rn}$  concentrations usually occur during spring and summer when convection and boundary layer mixing are most active at inland surface sites (Wilkening, 1959; Lindeken, 1967). Higher wintertime concentrations at central European sites were also likely attributed to slow transport and long residence time overland due to air

mass stagnation (Chambers et al., 2016a; Williams et al., 2016). At mid-latitude sites, snow cover suppresses  $^{222}\text{Rn}$  exhalation and reduces emission fluxes substantially in winter; complete snow melt and moist flux enhance  $^{222}\text{Rn}$  emissions in summer (Reithmeier and Sausen, 2002). Since strong emissions in summer partially compensate the dilution effect of boundary-layer mixing and strong convection, the lowest  $^{222}\text{Rn}$  concentrations are usually observed in the springtime. All simulations capture the seasonal variations; ZK11 and ZKC emission scenarios do not lead to obviously better results than JA97 and SW98. It appears that a sharp increase in emission is missing from summer to late fall as indicated by increased observations in June-August, suggesting that further emission adjustments are needed for Europe in the model. Szegvary et al. (2009) also suggested large  $^{222}\text{Rn}$  emissions over the Iberian Peninsula and the northern Mediterranean coastal region due to a wide coverage by dry soil and crystalline rocks. In a more recent study using  $^{222}\text{Rn}$  as a tracer to classify atmospheric stability in Slovenia, unusually large  $^{222}\text{Rn}$  exhalation flux from flysch and carbonate rocks at an inland site was found to cause higher  $^{222}\text{Rn}$  concentrations in the diurnal cycle compared to a coastal site where atmospheric synoptic conditions were considered similar but land was more dominated by sea and lake sediments (Kikaj et al., 2019).

Figure 8g shows the model-observation comparison for Mace Head, a coastal site in Western Europe (Ireland). Most observations are lower than  $1000 \text{ mBq SCM}^{-1}$  with a weak seasonal variation. Simulations with JA97 and SW98 overestimate the observations with fluctuated seasonality by more than 2 times a factor of >2 on average, while such large overestimates are only showsseen in February for ZK11 and ZKC. The coastal site is usually moist and largely affected by oceanic air; it is therefore characterized by relatively low  $^{222}\text{Rn}$  concentrations all year round. A regional model simulation by Chevillard et al. (2002) with a JA97-like, uniform  $^{222}\text{Rn}$  emission rate, showed similar overestimation with much larger discrepancies from observations. The site is located ( $53.3^\circ\text{N}$ ) very close to the cut-off latitude ( $60^\circ\text{N}$ ) in JA97, at which zero emissions are assumed northwards. The comparisons in Europe suggest that the fixed emission fluxes (with reductions under freezing conditions) in JA97 can lead to overestimation in southern Europe, underestimation in the north, and a weaker latitudinal gradient towards the north as shown by

ZK11 and ZKC is much favored. The comparisons with measurements applied with scaling factors suggested by Schmithüsen et al. (2017) are given in Fig. S3, which only shows slight changes.

Asia. Observations of surface  $^{222}\text{Rn}$  concentrations in Asia, e.g., southern China (Zahorowski et al., 2005), Japan (Chambers et al., 2009; Iida et al., 2000), and India (Debaje et al., 1996), are clearly affected by the Asian summer monsoon, with maximum concentrations observed in winter and minimums in summer (low- $^{222}\text{Rn}$  marine air brought by the monsoon). Figure 9 shows the model-observation comparisons at five Asian sites (Beijing, Gosan, Fuzhou, Hong Kong, and Bombay). Inland sites in China, where only annual mean observations are available, are not included in this comparison. The observations at Beijing show a moderate seasonal variation similar to the mid-latitude continental European sites, with a spring minimum and an autumn maximum. The simulation with JA97 shows reasonable agreement with observations at Beijing only in spring and summer, but is significantly biased low in late fall-early spring (November-March, Fig. 9a). The latter is likely due to the temperature-dependent reduction of  $^{222}\text{Rn}$  emissions in JA97 when surface temperature is below  $0^\circ\text{C}$ . In reality, soil may not be frozen when temperature remains below  $0^\circ\text{C}$  for a short period of time. At Gosan, an island site largely affected by Asian monsoon and emissions from the major Asian continent, observations show a strong seasonal variation with a winter maximum and a summer minimum. The large winter low bias at Gosan with JA97 is likely also due to the assumed dependency on surface temperature.

At two southeastern China coastal sites, Fuzhou and Hong Kong, the model-model results ~~at the corresponding grid-boxes largely underestimates~~ are much lower than the observations ~~for comparison~~ (Fig. 9c and 9d) (Fig. S4). ~~We used the model results in the grid boxes to the west of the ones where the sites are actually located (Fig. 9c and 9d) because the observations at the two sites more represent air masses of land origin and grid boxes are dominated by water due to the coarse resolution of the model. We tried sampling the model results at adjacent gridboxes and found that those for the gridbox to the west are much more comparable to the observed (Fig. 9c and 9d). This suggests that the observations at both sites are significantly affected by local  $^{222}\text{Rn}$  emissions.~~ The  $^{222}\text{Rn}$  observations show a minimum in summer,

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reflecting the intrusion of low-<sup>222</sup>Rn marine air associated with the Asian summer monsoon. Although the model successfully captures the observed seasonality-variation, the simulation with ZKC (with enhanced emissions in China) shows a much better agreement compared to the underestimated concentrations~~large low bias in the simulation with JA97. all simulations underestimate observed values all year round,~~ especially at Fuzhou (Fig. 9c). ~~The simulation with ZKC (with enhanced emissions in China) also results in large underestimation. This is likely attributed to the unusually high emission fluxes in southeastern China possibly due to the rocky texture in the mountainous region.~~ On the other hand, the simulations with ZK11 and ZKC capture the observations at Bombay, India, well. These contrasting model performances suggest that <sup>222</sup>Rn emission fluxes in southeastern China need to be better quantified with flux measurements at more surface sites.

**North America.** Figure 10 shows the model-observation comparisons at four U.S. continental sites. Similar to those mid-latitude surface sites in Europe and Asia, the observations at the U.S. sites show seasonal lows in spring and highs in fall or winter. The simulations with SW98 and ZKC (identical emissions over North America) show much higher <sup>222</sup>Rn concentrations than those with JA97 and ZK11 over the U.S. The seasonality at Chester is well captured by using SW98 and ZKC. At Cincinnati, the model performs slightly better with JA97 and ZK11, while the simulations with SW98 and ZKC overestimate the autumn peaks by nearly a factor of two. SW98 and ZKC lead to significant positive biases at Washington D.C., even though ZK11 commits negative biases of a similar magnitude. At Socorro, an elevated site (1400 m a.s.l.) in southern U.S., all simulations hardly do not convincingly capture the seasonal variation (Fig. 10d). Socorro is located in the Rio Grande Valley, where <sup>222</sup>Rn emissions may have larger variations due to surface textures and local meteorology (e.g., upslope air flows) that cannot be resolved by the coarse resolution model.

**Other Sites.** Figure 11 shows the seasonal variations of surface <sup>222</sup>Rn concentrations at eight sites in remote areas or the Southern Hemisphere. Surface <sup>222</sup>Rn concentrations at Bermuda show a late spring to summer minimum (May-August) due to the strengthened Azores-Bermuda High pressure system in



atmosphere. The sparse measurements ~~on~~at the edge of the Antarctic are not adequate ~~enough to help us for~~  
inferring emission fluxes ~~draw a flux map for~~over the remote continent. More future measurements of  
<sup>222</sup>Rn emissions in Antarctic regions are thus desired.

## 5 6 Vertical distribution of <sup>222</sup>Rn concentrations

The vertical distribution of <sup>222</sup>Rn reflects mainly the convective transport process rather than large-  
scale advection due to the relatively short decay lifetime (a few days) of <sup>222</sup>Rn. However, it is more  
challenging for global models to capture the convective transport of <sup>222</sup>Rn concentrations to the middle and  
upper troposphere than the synoptic-scale transport (Jacob et al., 1997). In this section, we characterize the  
convective transport in GEOS-Chem driven by the MERRA and GEOS-FP meteorological data sets,  
10 respectively, and evaluate model simulations with observed <sup>222</sup>Rn vertical profile.

### 6.1 Simulated <sup>222</sup>Rn profiles and comparison with observations

The most widely used <sup>222</sup>Rn profile measurements were compiled by Liu et al. (1984) (black line, Fig.  
12a). The composed profile is averaged from <sup>222</sup>Rn observations over the U.S., Ukraine, and central Asia  
15 and represents the summer <sup>222</sup>Rn vertical distributions over northern mid-latitude continental regions. The  
profile shows an inflection point between 3 and 4 km, reflecting the average altitude of convective  
entrainment (Fig. 12a). Concentrations decrease slowly as altitude increases from 4 to 7 km, indicating fast  
convective transport over land during summer (Liu et al., 1984; Zhang et al., 2008). We sample the  
simulated monthly mean <sup>222</sup>Rn profiles at the provinces or states where each observed profile was  
20 measured, and obtain an average profile for each simulation. As shown in Fig. 12, all simulations well  
capture the rapid decrease of <sup>222</sup>Rn concentrations from the surface to about 4 km at a rate of 1000 *mBq*  
*SCM<sup>-1</sup>* per *km*. The simulated concentrations then decrease faster than the observations until 6 km. It is  
suggested to be a consequence of overly vigorous convective transport in the model with detrainment at  
overly high altitudes (Considine et al., 2005). MERRA exhibits a higher and deeper convection from 5 to  
25 10 km. As a result, a remarkable underestimation of <sup>222</sup>Rn concentrations with MERRA is seen from 4 to 8

km, followed by overestimations above 9 km. ~~Overshot cloud top height for deep convection~~ Deep convective cloud top in MERRA has been seen in a comparison shown biased high with ~~compared to CERES-observed clouds~~ (Posselt et al., 2012). Stanfield et al. (2019) found that the frequency distribution of convective entrainment rates (mixing between environmental air with in-cloud air) for deep convection events in GEOS-5 has a significantly larger fraction in the higher-end values compared to the rates derived from TES/MLS-observed CO profiles. Intensive mixing ~~during~~ within convective updrafts undermines the upward lifting of surface air masses to the upper troposphere, possibly causing the rapidly decreasing <sup>222</sup>Rn concentrations with height in the simulation with GEOS-FP ~~driven simulation~~. Due to weaker convection in GEOS-FP, the simulation underestimates in a broader altitude range (4-10 km). It seems challenging for the two GEOS products to capture the convective detrainment level. As pointed out below, weaker convection in GEOS-FP at the resolution of  $2^\circ \times 2.5^\circ$  is partially due to the transport errors introduced by using the archived and regridded meteorological data (Yu et al., 2018).

Figure 12b compares model results with the <sup>222</sup>Rn profile averaged from measurements obtained at Moffett Field, a coastal site in California, U.S., during June to August in 1994 (Kritz et al., 1998). The model profiles are obtained by averaging monthly <sup>222</sup>Rn concentrations in the grid column corresponding to the site and those in the grid column to the west as suggested by Zhang et al. (2008). The simulations ~~hardly marginally~~ capture the “C” shape profile, a sign of strong convective transport in summer. The simulation with JA97 performs better until up to 5 km, above which those with ZKC and SW98 agree better with the observations. The large overestimation at 2 to 5 km with ZKC and SW98 is likely due to too strong shallow convection and/or high emission fluxes. The differences in near-surface concentrations between the simulations with ZKC and SW98 (Fig. 12b) are caused by averaging ZK11 and SW98 emission fluxes along the edges of the continent in the formulation of ZKC.

Figure 12c shows the comparison of model simulations with the profile averaged from aircraft measurements in the east coastal region of Canada during NARE in August 1993 (Zaucker et al., 1996). The model results are averages over a region of 41-46°N and 60-70°W. The simulation with JA97

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reasonably reproduces the observations between 0-4 km, while the simulation with ZKC overestimates between 2 and 5 km. The model performance for NARE is similar to that for Moffett Field. The stronger emissions (ZKC and SW98) tend to result in overestimates in the lower free troposphere (Moffett Field and NARE) but better estimates in the upper troposphere (Moffett Field).

5 The vertical  $^{222}\text{Rn}$  profiles at Goulburn were measured up to about 3.5 km above the ground level in May of 2006-2008 (Fig. 12d; Williams et al., 2011). The corresponding model results are monthly means for May of the simulation year. The model underestimates the concentrations substantially but well simulates the vertical gradient. It suggests that the underestimation is more likely caused by potentially low biases in the emissions over the Australian continent rather than errors associated with vertical mixing in  
10 the model. Despite this, the model reproduces well the seasonality in surface  $^{222}\text{Rn}$  observations at Cape Grim (Fig. 11f), which is located on the Tasmania Island to the south of the Australian continent. Above 2.5km, the vertical gradient of  $^{222}\text{Rn}$  concentrations decrease in both the observations and the model.

Two model uncertainties may affect our simulated  $^{222}\text{Rn}$  profiles: the remapping of the meteorological data from the original cubed-sphere grid in the parent GCM (GEOS-5) to an equally rectilinear (latitude-  
15 longitude) grid in the off-line CTM (GEOS-Chem); the degradation of the temporal and spatial resolutions of the meteorological archive (Yu et al., 2018). Yu et al. demonstrated that such remapping and using 3-hourly averaged wind archives may introduce 5-20% low biases into vertical transport of  $^{222}\text{Rn}$ , including the weakened transport from the boundary layer to the upper troposphere. They also showed that degrading the spatial resolution of the meteorological archive for input to GEOS-Chem further weakened vertical  
20 transport because organized vertical motions in the finer resolution are averaged out in the coarser resolution. Such biases may partially contribute to the discrepancies between the simulated and observed  $^{222}\text{Rn}$  profiles, which appear to be larger in the mid- and upper- troposphere (5.5-10 km) when the model is driven by GEOS-FP (Fig. 12). GEOS-FP has finer native horizontal resolution (0.25° latitude by 0.3125° longitude) than MERRA reanalysis (0.5° latitude by 0.667° longitude), [and exhibits weaker convection](#)  
25 [likely due to a more expanded intensive regriding.](#) An effort is currently ongoing to restore the lost

vertical transport by implementing the modified Relaxed Arakawa-Schubert convection scheme in GEOS-Chem (He et al., 2019).

## 6.2 Role of convective transport: MERRA vs. GEOS-FP

To examine the role of convective transport in simulated distributions of  $^{222}\text{Rn}$ , we compare model simulations driven by MERRA and GEOS-FP where the convective transport operator is turned on or off (i.e., A1, B1, A1-nc, and B1-nc, where “nc” denotes no convection; Table 24). Figure 13 shows the latitude-pressure cross-section of ~~annual~~-zonal mean  $^{222}\text{Rn}$  concentrations in these four simulations ~~for Northern Hemisphere~~ averaged over the boreal summertime (June, July, and August). The concentrations are contoured on a logarithmic scale. The strong gradients above the tropopause in all panels are indicative of a fast decrease of  $^{222}\text{Rn}$  concentrations due to weak vertical diffusion. The interhemispheric asymmetry in  $^{222}\text{Rn}$  distributions reflect the larger landmass and  $^{222}\text{Rn}$  emissions in the Northern Hemisphere. The latitudinal and vertical distributions of  $^{222}\text{Rn}$  concentrations simulated with MERRA and GEOS-FP are very similar. The overall vertical transport in the simulation with MERRA is slightly stronger than with GEOS-FP as shown by the higher  $^{222}\text{Rn}$  concentrations near the ~~sub-tropical tropopause~~ between 15-30°N (Fig. 13a-b). In contrast, when convection is turned off (Fig. 13c-d), the model simulates higher  $^{222}\text{Rn}$  concentrations near the tropical tropopause with GEOS-FP than with MERRA, indicating that convection is stronger in MERRA than in GEOS-FP.

Figure 14 shows the ~~relative contributions~~ percentage changes in  $^{222}\text{Rn}$  ~~annual~~-zonal mean concentrations ~~for Northern Hemisphere~~ averaged over the boreal summertime ~~due to~~ convection in MERRA and GEOS-FP, defined as  $(^{222}\text{Rn} - ^{222}\text{Rn}_{nc})/^{222}\text{Rn} \times 100\%$  where  $^{222}\text{Rn}$  and  $^{222}\text{Rn}_{nc}$  denote simulations with and without the convection operator, respectively. Where positive values occur, convection facilitates the transport of  $^{222}\text{Rn}$  to the region and increases  $^{222}\text{Rn}$  concentrations. Similarly, negative values indicate convection decreasing  $^{222}\text{Rn}$  concentrations. The negative values in the lower troposphere of the Northern Hemisphere along with the positives in the middle and upper troposphere are due to the pumping effect of convection, transport surface-emitted  $^{222}\text{Rn}$  upward. Convection in the

simulation with GEOS-FP transports about 20-30% less  $^{222}\text{Rn}$  to higher altitudes in the tropics and subtropics compared to MERRA (Fig. 14a vs 14b). Figure 15 shows the annual mean convective and large-scale vertical fluxes of  $^{222}\text{Rn}$  in the simulations with MERRA and GEOS-FP as well as their differences. Convective fluxes are stronger in a broader latitude range (30°S-55°N) in the simulation with MERRA. The largest difference appears in the tropical lower troposphere where convective fluxes of  $^{222}\text{Rn}$  in the simulation with MERRA are about a factor of two larger than those in the simulation with GEOS-FP (Fig. 15c). The large-scale vertical fluxes of  $^{222}\text{Rn}$  in the simulation with GEOS-FP are significantly larger than those with MERRA (Fig. 15f), partly compensating the differences in convective fluxes. This compensation leads to the aforementioned general similarity in the zonal mean  $^{222}\text{Rn}$  distributions in the two simulations (Fig. 13).

To further illustrate the differences in convective transport between the simulations with MERRA and GEOS-FP, we show in Fig. 16 the simulated  $^{222}\text{Rn}$  profiles averaged over the northern mid-latitude land areas (30°-60°N) for both cases of with and without convection. The solid black line with the upper x-axis presents the corresponding concentration ratios between the two simulations. Similar to the earlier analysis of  $^{222}\text{Rn}$  vertical fluxes, convection in MERRA is stronger as indicated by the large change in  $^{222}\text{Rn}$  concentrations at high altitudes (e.g., 8 km) when convection is off (solid red line vs. dashed red line). The different characteristics of vertical transport in MERRA and GEOS-FP are better revealed by examining the  $^{222}\text{Rn}$  concentration ratio profiles (black and green lines with the upper x-axis). Convective transport takes effect from the base of cloud layers (i.e., the lowest model layer with non-zero convective mass fluxes) in the model, whereas the large-scale vertical advection occurs from the bottom model layer up. As shown by  $^{222}\text{Rn}$  concentration ratios between the two simulations with convection turned off (green line, Fig. 16), ~~it is more efficient in GEOS-FP than in MERRA to transport  $^{222}\text{Rn}$  vertically through large-scale advection and boundary-layer mixing~~ the vertical transport of  $^{222}\text{Rn}$  through large-scale advection and boundary-layer mixing is more efficient in GEOS-FP than in MERRA ( $^{222}\text{Rn}$  ratios < 1 above ~2.5 km and > 1 below). Even with convection turned on, simulated ~~near-surface~~  $^{222}\text{Rn}$  concentrations ~~near the surface~~

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are still lower in GEOS-FP than in MERRA (solid black line, Fig. 16) because large-scale advection and boundary-layer mixing dominate near the surface and drain surface  $^{222}\text{Rn}$  faster. When  $^{222}\text{Rn}$  reaches the base of convective clouds, it is more efficiently uplifted in MERRA due to stronger convection, resulting in lower  $^{222}\text{Rn}$  concentrations in the lower troposphere ( $^{222}\text{Rn}$  ratios  $< 1$  from  $\sim 0.75$  to 4 km), and higher concentrations in the middle to upper troposphere ( $> 4$  km). This feature should also affect the simulations of other surface emitted species when using MERRA and GEOS-FP as the driving meteorology in GEOS-Chem.

## 7 Summary and conclusions

We have evaluated the global distributions of  $^{222}\text{Rn}$  simulated by the GEOS-Chem chemical transport model with a focus on the sensitivity of simulated surface concentrations and seasonality to the choice of available emission scenarios. A preferred emission scenario was recommended based on evaluations against surface observation of  $^{222}\text{Rn}$  concentrations and  $^{210}\text{Pb}$  deposition fluxes. We have discussed the major factors controlling  $^{222}\text{Rn}$  emissions as well as potential emission uncertainties in East Asia, North Africa, and Antarctic. We have also characterized the vertical transport processes associated with the MERRA and GEOS-FP meteorological data products by comparing simulated  $^{222}\text{Rn}$  vertical profiles with observations.

We implemented three new global  $^{222}\text{Rn}$  emission scenarios in GEOS-Chem, SW98 (Schery and Wasiolek, 1998), ZK11 (Zhang et al., 2011), and ZKC (an optimized inventory modified from Zhang et al. (2011)). All scenarios include prescribed regional variations and seasonality, which are lacking in the JA97 emission scenario (Jacob et al., 1997) currently used in the standard GEOS-Chem and other global models. JA97 often led to much larger biases in surface concentrations relative to the other scenarios because of lack of spatial variations and overly simplified emission reduction under freezing conditions (e.g., in high-latitude regions). The new emission options all resulted in remarkable increases in surface  $^{222}\text{Rn}$  concentrations at northern mid- and high-latitudes. Such increases were more pronounced in winter due to

the accumulation effect within the shallow boundary-layer. With constraints from observations, we are able to achieve much better agreements between the model and observations in all four defined regions (Europe, Asia, North America, and remote regions) using a customized emission scenario, ZKC. However, the simulation with ZKC still inherited some unsolved issues, e.g., large biases in Asia, poorly characterized emission fluxes in Antarctica, and at some elevated sites. More measurements of soil radium content and surface  $^{222}\text{Rn}$  concentrations are desired to produce a better global  $^{222}\text{Rn}$  emission scenario. The seasonality in surface  $^{222}\text{Rn}$  concentrations at northern mid-latitudes typically shows a low in spring and a peak in fall, a result of the competition between changes in emission fluxes and the strength of vertical transport (ventilation). In subtropical East and South Asia, the seasonality is strongly affected by monsoon and shows a summer minimum. Our analyses also suggested that  $^{222}\text{Rn}$  emissions have been quantified more accurately over Europe due to more frequent and evenly distributed measurements across the continent.

We specifically investigated the ~~excessive-underestimated~~ Asian  $^{222}\text{Rn}$  emissions and explored possible reasons based on previous studies. Both our simulated surface  $^{222}\text{Rn}$  concentrations and  $^{210}\text{Pb}$  (decay daughter of  $^{222}\text{Rn}$ ) deposition fluxes over Asia suggested ~~excessive-underestimated Asian~~  $^{222}\text{Rn}$  emissions in the model/Asia. In the simulation experiments with Asian  $^{222}\text{Rn}$  emissions scaled up by a factor of 1.2 to 1.7, agreements with surface observations ~~were-was~~ significantly improved. However, due to limited knowledge about the spatial distributions and extents associated with the underestimation in Asian emissions, we did not apply a larger scaling factor, which would cause large overestimates of  $^{210}\text{Pb}$  deposition fluxes in the model. As a trade-off, we used a scaling factor of 1.2 for emissions over China in the ZKC inventory, which increased the simulated surface  $^{222}\text{Rn}$  concentrations and led to a better agreement with observations in Asia. The issue of underestimated Asian emissions is still open. An ideal solution would be an improved and spatially resolved emission map instead of using a uniform scaling factor for the region. The excessive  $^{222}\text{Rn}$  emissions in Asia may be due to multiple factors, including various surface textures, high contents of radium in the soil, active crust movement along the Asian earthquake zone, and high contents of radium in the fertilizer used in East Asia and India.

We found that it was challenging for model simulations driven by GEOS products to fully capture the vertical structure of observed  $^{222}\text{Rn}$  profiles. A comparison with summertime continental profiles showed that both MERRA and GEOS-FP have biased levels of convective detrainment. Convection in both MERRA and GEOS-FP was likely too deep in northern mid-latitude land areas. ~~The strength of convection in GEOS-FP is too weak, leading to large low biases of  $^{222}\text{Rn}$  in the mid-high troposphere. The weak convection in GEOS-FP leads to large low biases of  $^{222}\text{Rn}$  in the mid-high troposphere.~~ This is partly attributed to the lost vertical transport as a result of the remapping from the cubed-sphere to equally rectilinear grids and the degradation of the spatiotemporal resolution of the input meteorological data (Yu et al., 2018). A comparison of global  $^{222}\text{Rn}$  vertical distributions between the simulations driven by MERRA and GEOS-FP showed a distinct difference in the role of convective transport (versus large-scale vertical advection) in determining the  $^{222}\text{Rn}$  vertical distributions. The stronger convective transport in MERRA is partially compensated by its weaker large-scale upward advection compared with GEOS-FP, resulting in similar vertical  $^{222}\text{Rn}$  distributions in the model simulations driven by the two meteorological products. This has important implications for using chemical transport models to interpret the transport of other trace gases and aerosols when these GEOS products are used as driving meteorology.

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*Data availability.* The  $^{222}\text{Rn}$  emission data used in this paper is described in Section 2.2. Observational data for model evaluation are introduced in Section 2.3. All model output, emission data, and observational data sets are available online (<http://doi.org/10.5281/zenodo.3942287>).

*Author contribution.* BZ and HL designed the study. BZ conducted the model simulations and led the analysis. BZ and HL wrote the manuscript with contributions from all coauthors. SC, CHK, and AGW contributed  $^{222}\text{Rn}$  surface and profile data sets. KZ contributed the ZK11  $^{222}\text{Rn}$  emission data and surface observational data sets. DBC contributed to the analysis of  $^{222}\text{Rn}$  vertical profiles. MPS and RMY contributed to the model development.

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**Tables and figures**

5 **Table 1. Global <sup>222</sup>Rn emission scenarios used in this work.**

Scenario	Reference	Description
JA97	Jacob et al. (1997)	Emission fluxes are 1.0 atom cm <sup>-2</sup> s <sup>-1</sup> over land between 60°N – 60°S, 0.005 atom cm <sup>-2</sup> s <sup>-1</sup> between 60°N – 70°N and 60°S – 70°S, zero poleward of 70°N/S, and 0.005 atom cm <sup>-2</sup> s <sup>-1</sup> over lakes and oceans. Emissions are reduced by a factor of 3 when surface temperature is below 0°C.
SW98	Schery and Wasiolek (1998)	Emission fluxes are formulated by using a theoretical diffusion model of porous soil with controlling factors of soil radium content, soil moisture, and surface temperature. Emission fluxes in SW98 were found to be overestimated and are reduced by a factor of 1.6 globally in this work (Koch et al., 2006, Zhang et al., 2011)
ZK11	Zhang et al. (2011)	Based on SW98, ZK11 updated emission fluxes over Europe, U.S., China, Australia, and oceanic regions according to more recent measurements.
ZKC	This work.	ZKC increases emission fluxes <u>over in the geographic territory of China</u> by a factor of 1.2 upon ZK11 and retrogresses to SW98 over U.S.

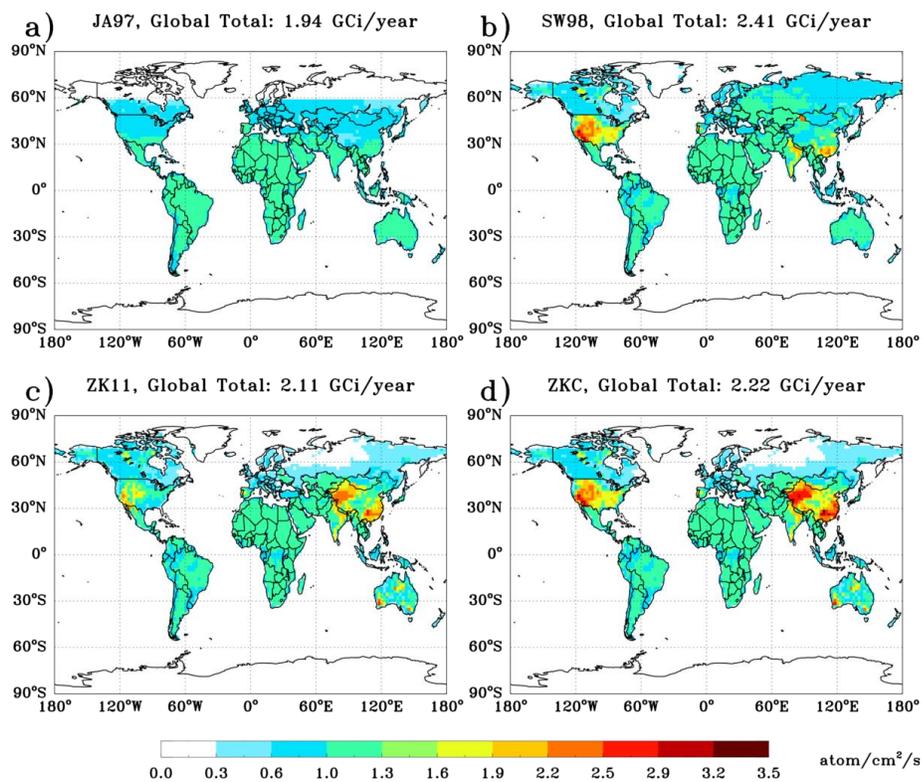
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**Table 21. Configurations of GEOS-Chem simulations (v11-01f, 2°×2.5°) used in this work.**

Simulation	<sup>222</sup> Rn emission	Driving Meteorology	Convection
A1	JA97 (Jacob et al., 1997)	MERRA	on
A2	SW98 (Schery and Wasiolek, 1998)	MERRA	on
A3	ZK11 (Zhang et al., 2011)	MERRA	on
A4	ZKC (this work)	MERRA	on
B1	JA97 (Jacob et al., 1997)	GEOS-FP	on
A1-nc	JA97 (Jacob et al., 1997)	MERRA	off
B1-nc	JA97 (Jacob et al., 1997)	GEOS-FP	off



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Figure 1. Global annual mean surface  $^{222}\text{Rn}$  emission fluxes ( $\text{atom cm}^{-2} \text{s}^{-1}$ ) of four emission scenarios used in this study. a) JA97: the default emission in the standard version of GEOS-Chem (Jacob et al., 1997); b) SW98: the first global  $^{222}\text{Rn}$  emission with regional variability based on a soil emission model (Schery and Wasiolek, 1998); c) ZK11: a recently published global  $^{222}\text{Rn}$  emission combining SW98 and recent measurements of  $^{222}\text{Rn}$  fluxes (Zhang et al., 2011); and d) ZKC: ZK11 with customized adjustments to better match observations (this work).

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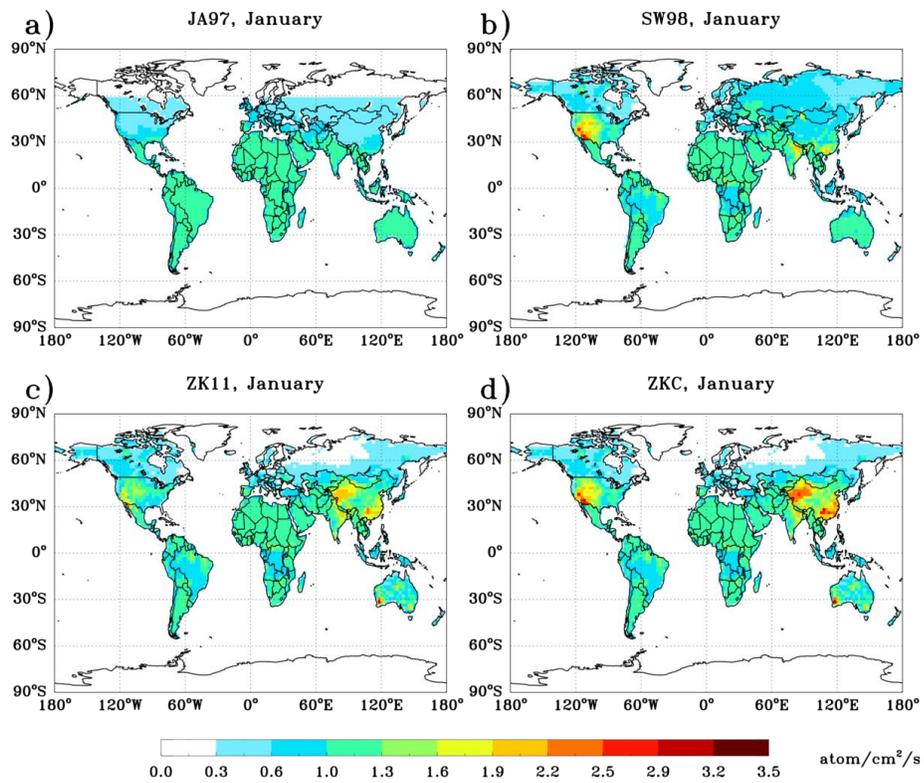


Figure 2. Same as Figure 1 but for January.

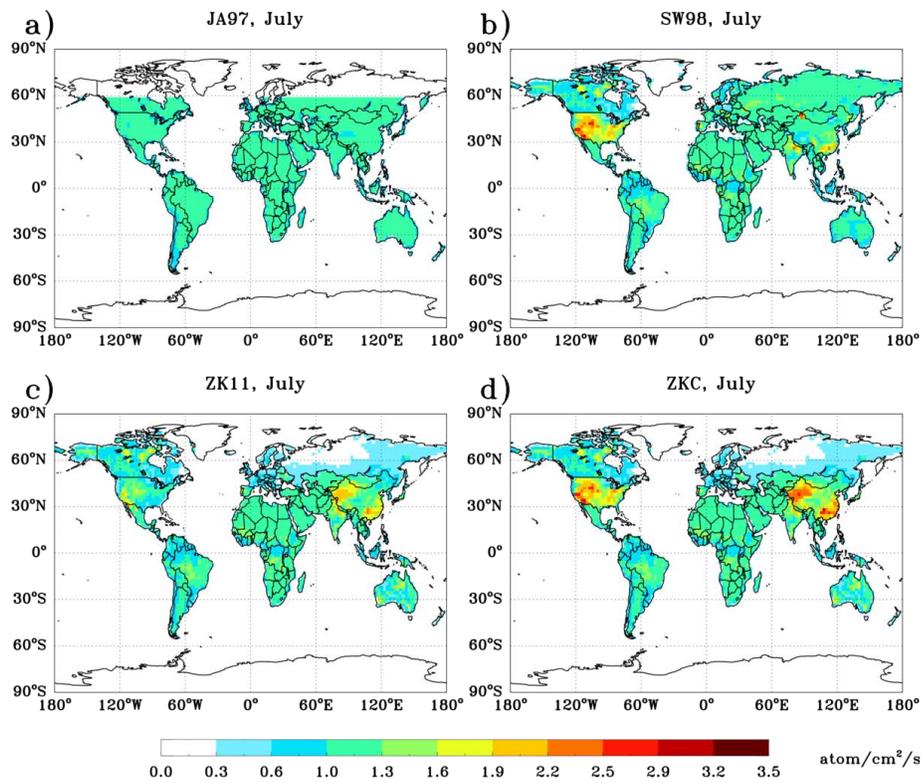
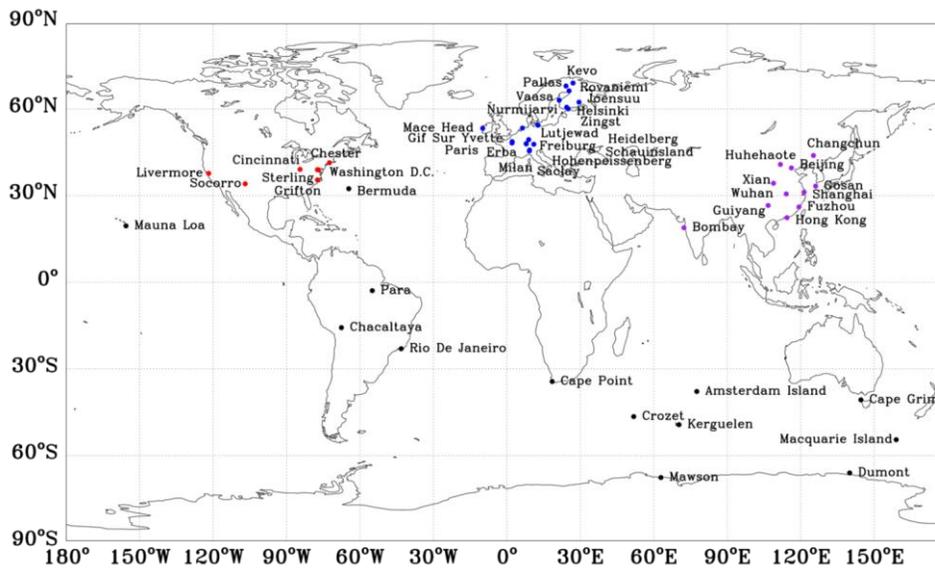


Figure 3. Same as Figure 1 but for July.



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Figure 4. Locations of surface  $^{222}\text{Rn}$  measurement sites. Sites in four distinctive regions are color-coded: Europe (blue), Asia (purple), North America (red), and remote regions (black). Refer to Table 2 of Zhang et al. (2011) for more details.

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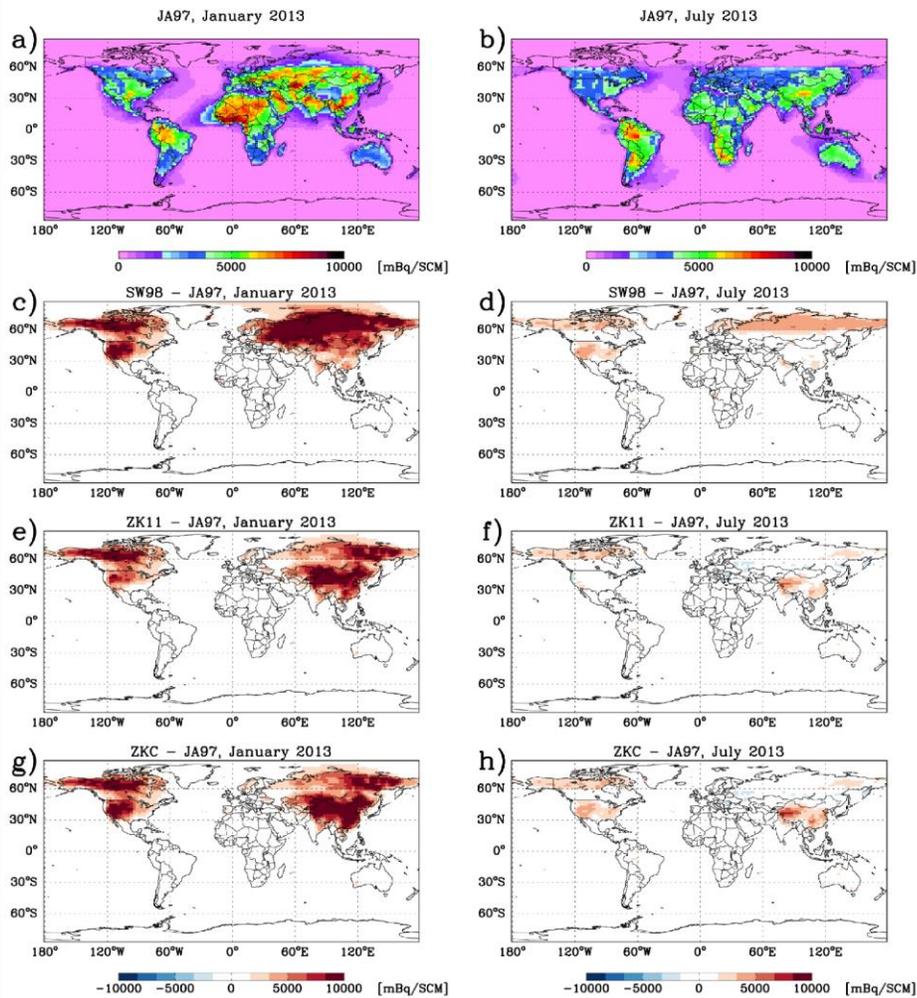


Figure 5. Simulated monthly surface  $^{222}\text{Rn}$  concentrations (mBq/SCM) for (a) January 2013 and (b) July 2013 with the JA97 emission scenario (A1, see Table 24). Panels (c)-(h) are same as (a) and (b) but showing the changes in surface  $^{222}\text{Rn}$  concentrations when SW98 (A2), ZK11 (A3), and ZKC (A4) emissions are used in the model, respectively.

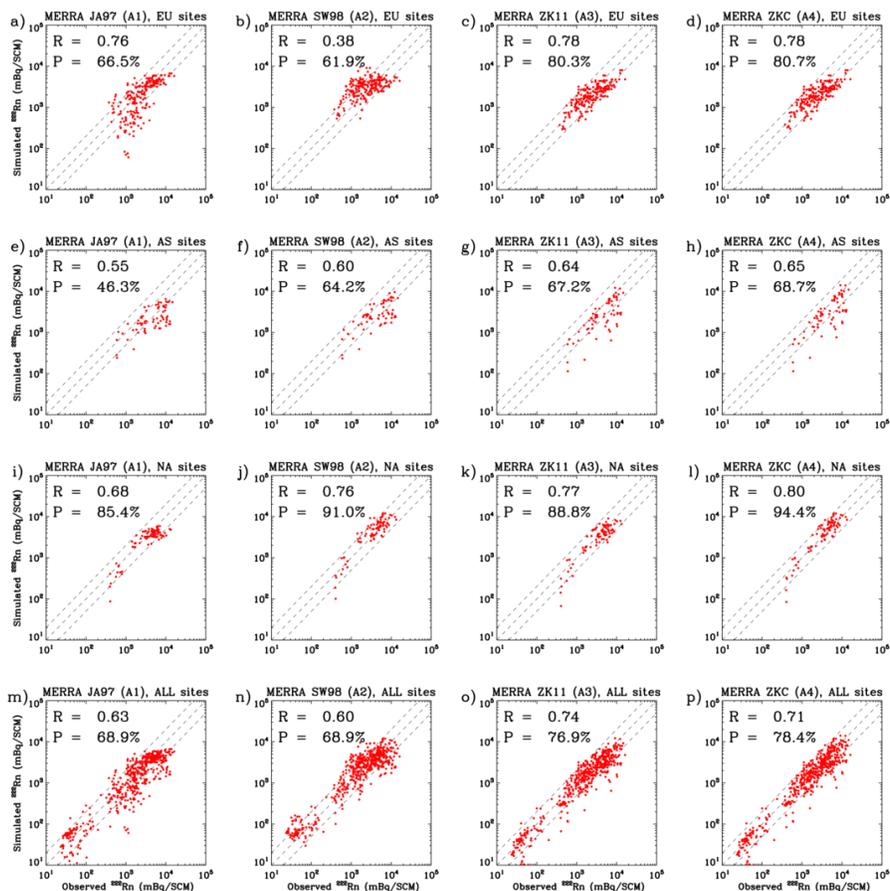


Figure 6. Comparisons between simulated and observed monthly surface  $^{222}\text{Rn}$  concentrations (mBq/SCM) over the continents of Europe (EU, first row), Asia (AS, second row), North America (NA, third row), and over the globe (ALL, last row), respectively. The four columns correspond to simulations (A1-A4) with the four emission inventories (JA97, SW98, ZK11, and ZKC, see Table 24). Dashed lines indicate the range within a factor of two of the 1 to 1 line. P is the percentage of samples within this range. R in the legends is the 2-sided linear regression correlation coefficient. The lines of best fit are calculated using the reduced-major-axis method (Hirsch and Gilroy, 1984).

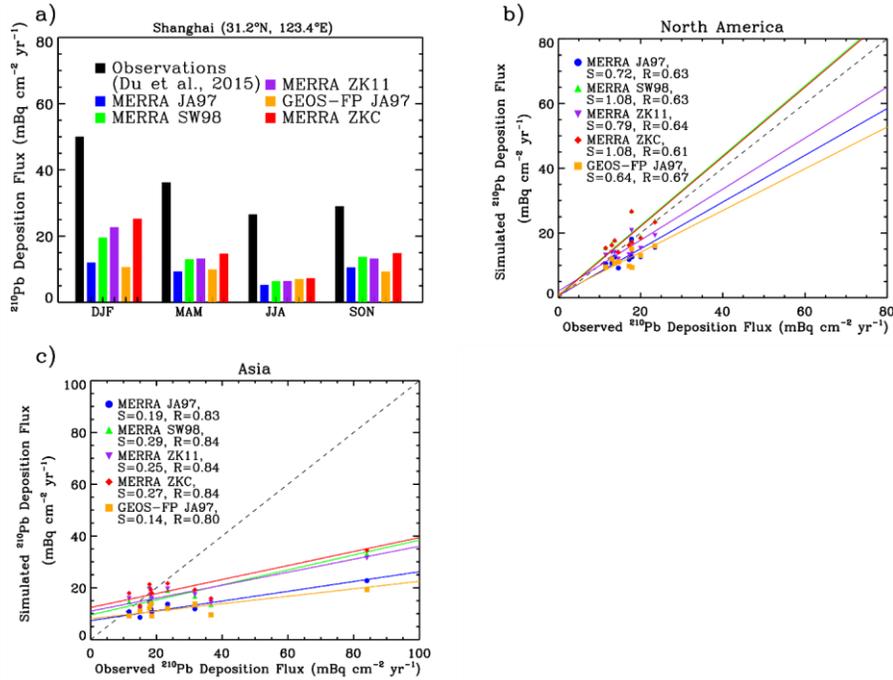


Figure 7. a) Comparison of seasonal total  $^{210}\text{Pb}$  deposition fluxes (mBq cm<sup>-2</sup> yr<sup>-1</sup>) at Shanghai (32.1°N, 123.4°E) between five model simulations (see Table 24) and observations (Du et al., 2015). b) Correlations between simulated and observed annual mean  $^{210}\text{Pb}$  deposition fluxes at nine surface sites in North America (Du et al., 2015). c) Same as b) but for nine Asian sites. Dashed line is the 1 to 1 line. Color lines are linear regression lines for the five model simulations shown in the legends. The reduced-major-axis regression slopes (S) and correlation coefficients (R) are given in the legends.

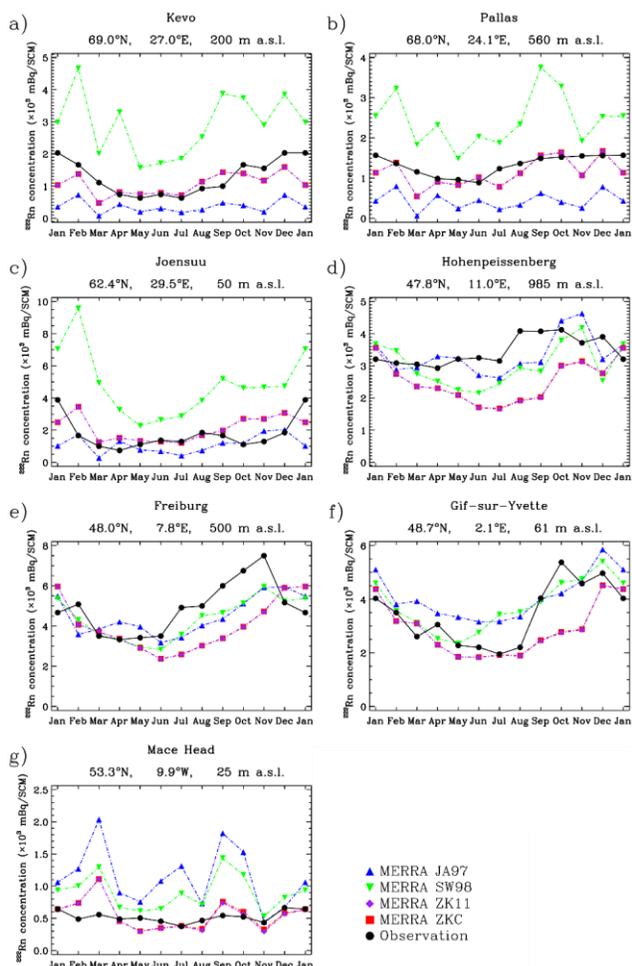
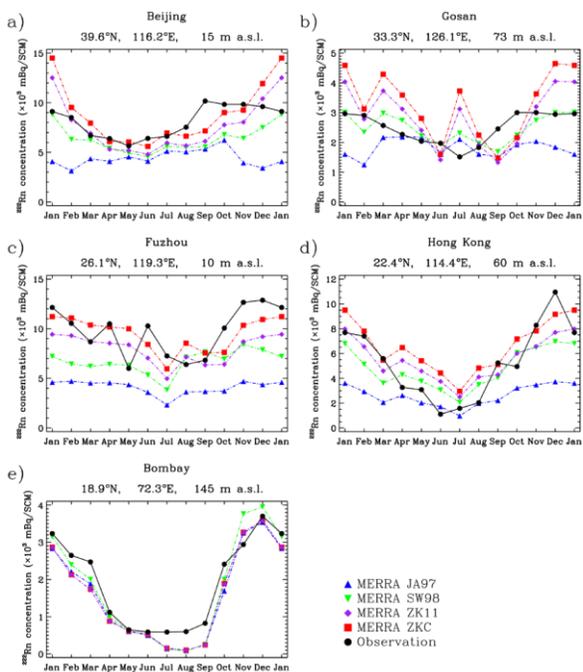


Figure 8. Comparison between simulated (color lines) and observed (black lines)  $^{222}\text{Rn}$  climatological monthly means (black lines)  $^{222}\text{Rn}$  concentrations (mBq/SCM) and simulated monthly means in 2013 (color lines) at selected surface sites in Europe. Location and elevation of each site are given upon each panel. See Table 24 for the list of model simulations. Note the small difference between the simulations with ZK11 and that with ZKC because of identical  $^{222}\text{Rn}$  emission in Europe.



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Figure 9. Same as Fig. 8, but for Asia. Please note that the model results used in panel c) Fuzhou and d) Hong Kong are sampled from the grid-boxes to the west of the ones where the stations sites are located for to achieve a better agreement with the observations. The observations at these two sites are likely more representative for air masses of land origin, whereas the model grid boxes are dominated by water. See text for details.

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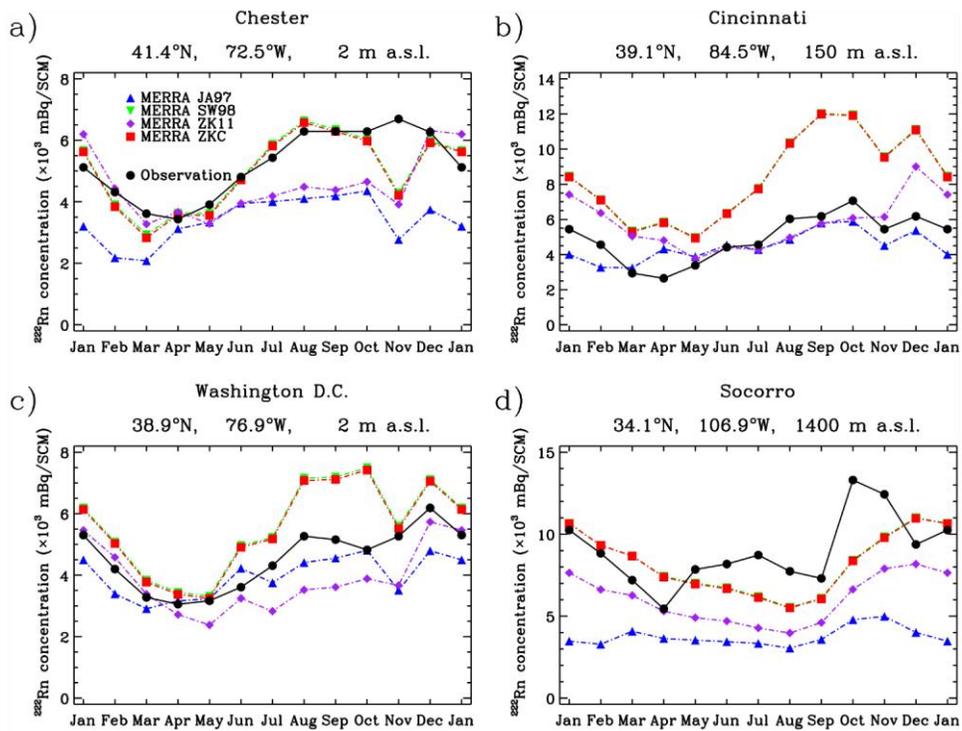


Figure 10. Same as Fig. 8, but for North America.

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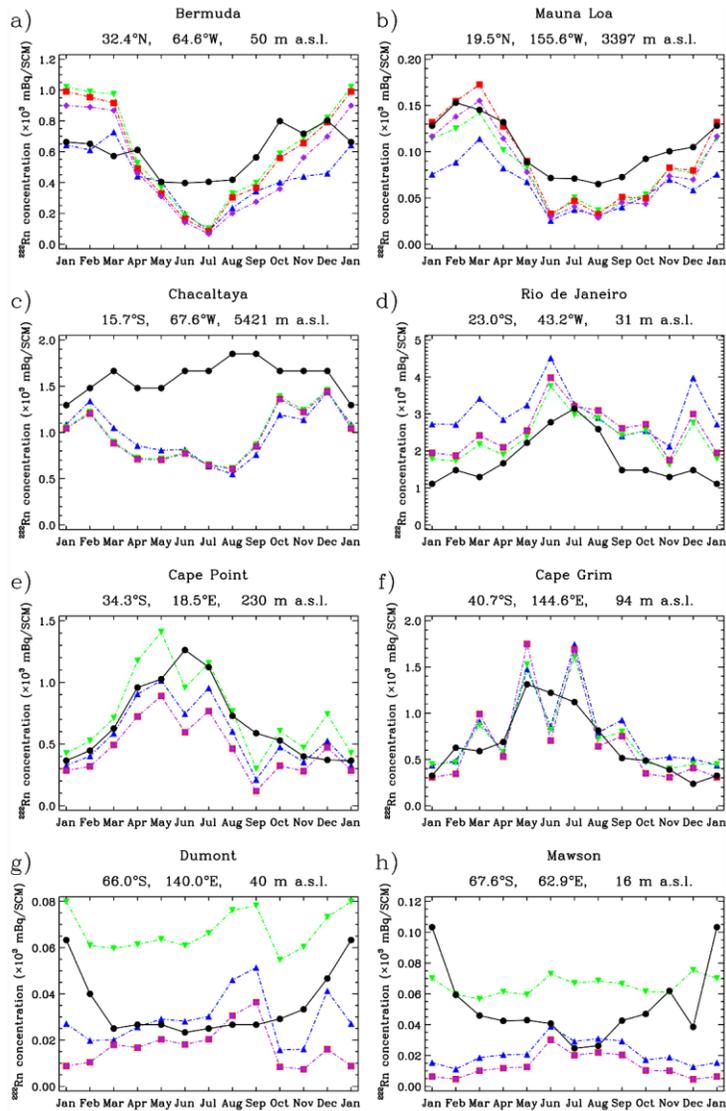


Figure 11. Same as Fig. 8, but for remote sites.

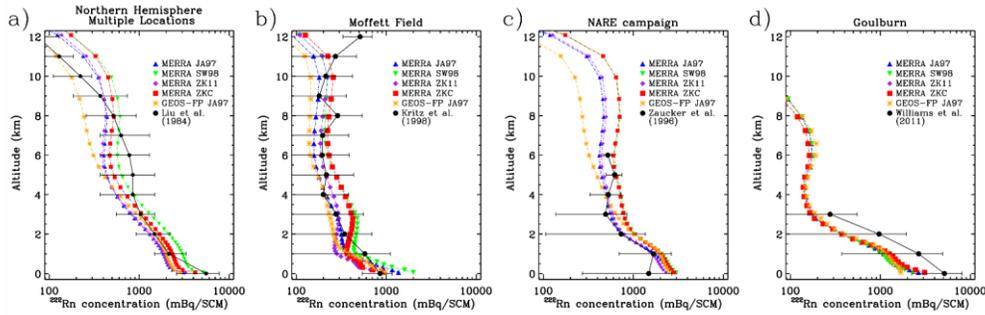


Figure 12. Comparison of vertical of  $^{222}\text{Rn}$  profiles (mBq/SCM) simulated with four emission scenarios (simulations A1, A2, A3, and A4, see Table 2+) with a) an average profile compiled from [multiple-23](#) locations over the Northern Hemisphere continents (Liu et al., 1984), b) an average summertime profile constructed from measurements at Moffett Field (37.4°N, 122°W), California (Kritz et al., 1998), c) an average summertime profile from measurements on the east coast of Canada during the 1993 NARE campaign (Zaucker et al., 1996), and d) an averaged profile measured in May of 2006–2008, at Goulburn (34.8°S, 149.7°E), New South Wales, Australia (Williams et al., 2011). In panel (a), more than half of the observed profiles reach up to 6–12km. Horizontal bars indicated the standard deviations of the observed  $^{222}\text{Rn}$  concentrations.

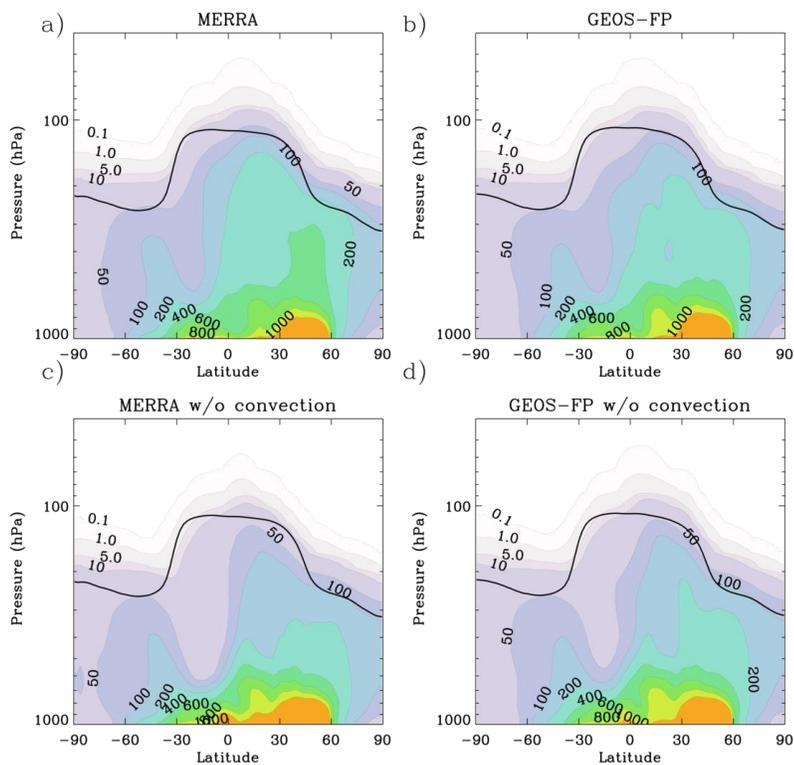


Figure 13. Latitude-pressure cross-sections of annual-zonal mean  $^{222}\text{Rn}$  concentrations averaged for over June-July-August (mBq/SCM) as simulated by the GEOS-Chem model driven by a) MERRA (A-1), b) GEOS-FP (B-1), c) MERRA without convection (A1-nc), and d) GEOS-FP without convection (B1-nc). Bold black lines denote the annual-zonal mean tropopause height (hPa) in the corresponding meteorological data set.

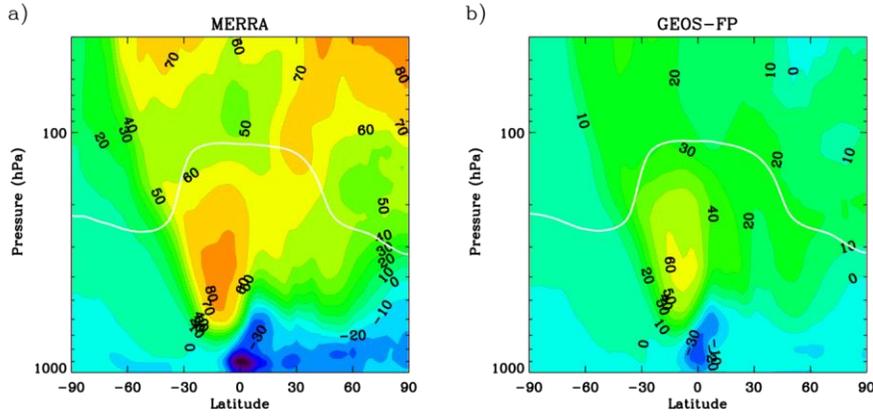


Figure 14. ~~Percentages~~ Percentage changes in of annual ~~June July August~~ zonal mean  $^{222}\text{Rn}$  concentrations averaged over ~~June-July-August~~ contributed by due to convective transport in the GEOS-Chem simulations driven by a) MERRA and b) GEOS-FP. Values are  $(^{222}\text{Rn} - ^{222}\text{Rn}_{nc}) / ^{222}\text{Rn} \times 100$ , where  $^{222}\text{Rn}$  and  $^{222}\text{Rn}_{nc}$  are  $^{222}\text{Rn}$  concentrations simulated with (A1 and B1, Table 24) and without (A1-nc and B1-nc) the convection operator, respectively.

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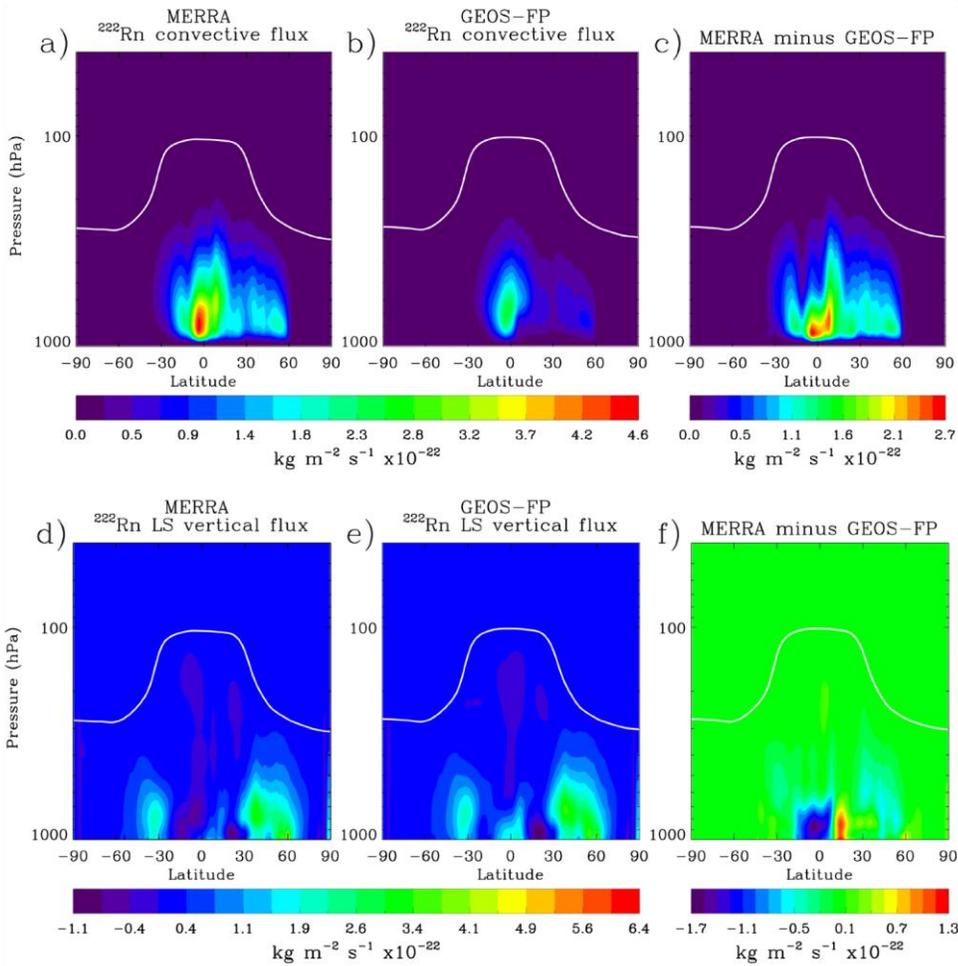


Figure 15. Comparison of annual zonal mean vertical fluxes of  $^{222}\text{Rn}$  ( $\times 10^{-22} \text{ kg m}^{-2} \text{ s}^{-1}$ ) in the GEOS-Chem simulations driven by MERRA and GEOS-FP. Upper panels: a) convective fluxes with MERRA, b) convective fluxes with GEOS-FP, and c) difference between a) and b). Lower panels: d) large-scale (LS) vertical fluxes with MERRA, e) large-scale vertical fluxes with GEOS-FP, and f) the difference between e) and f). The white lines indicate the tropopause height (hPa) in MERRA.

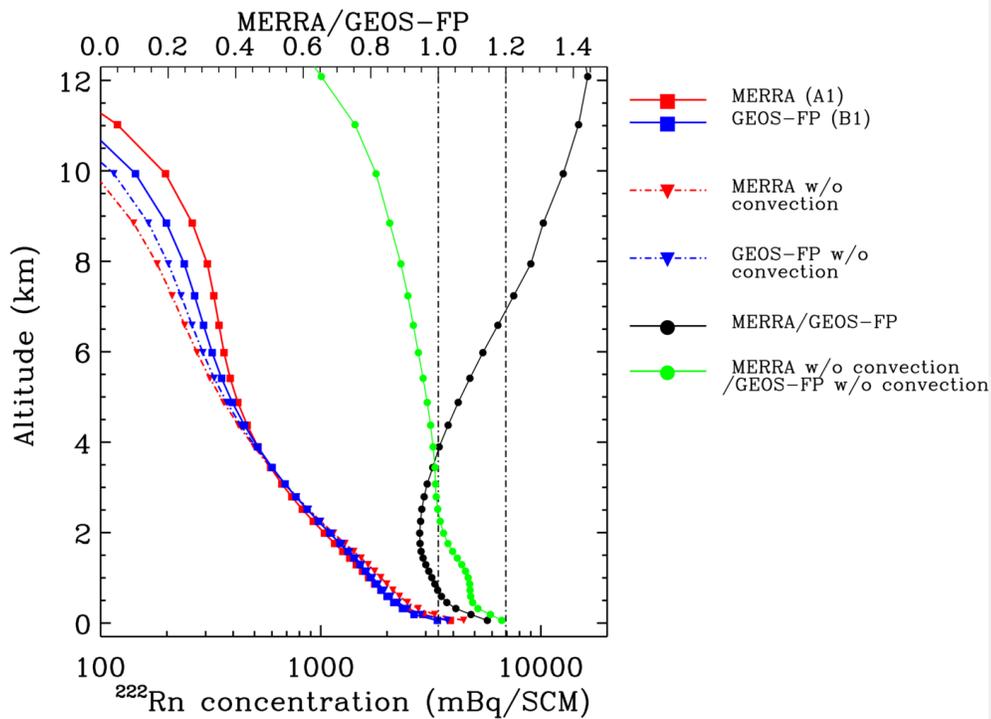


Figure 16. Annual zonal mean  $^{222}\text{Rn}$  profiles (mBq/SCM, red and blue lines) averaged over land areas between 30-60°N latitudes in simulations driven by MERRA (A1 and A1-nc, Table 2+) and GEOS-FP (B1 and B1-nc, Table 2+), respectively. The black solid line (with the upper axis) shows the ratios of simulated  $^{222}\text{Rn}$  concentrations in the standard simulations with MERRA and GEOS-FP. The green line shows the same ratios when convection is turned off in the simulations. The two black dot-dashed lines have constant ratios of 1.0 and 1.2, respectively.

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## *Supplement of*

**Simulation of radon-222 with the GEOS-Chem global model:**

**Emissions, seasonality, and convective transport**

5

Bo Zhang, Hongyu Liu, James H. Crawford, Gao Chen, T. Duncan Fairlie, Scott Chambers,  
Chang-Hee Kang, Alastair G. Williams, Kai Zhang, David B. Considine, Melissa  
P. Sulprizio, and Robert M. Yantosca

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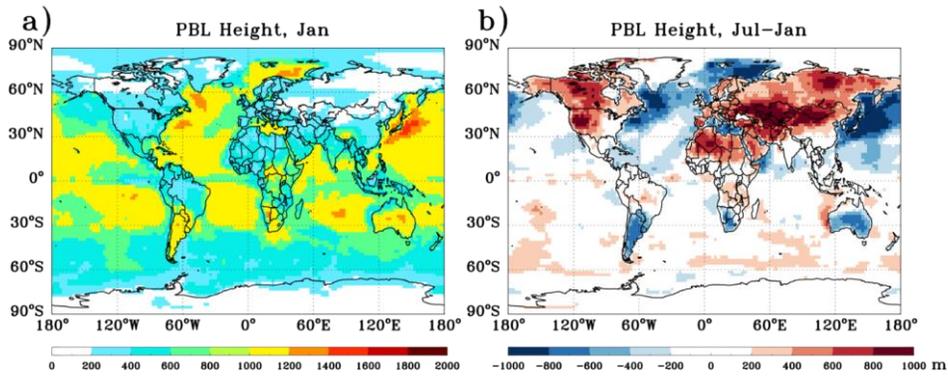
Manuscript submitted to ACPD, July 2020; [Revised Dec. 2020](#)

*Correspondence to:* Hongyu Liu (hongyu.liu-1@nasa.gov)

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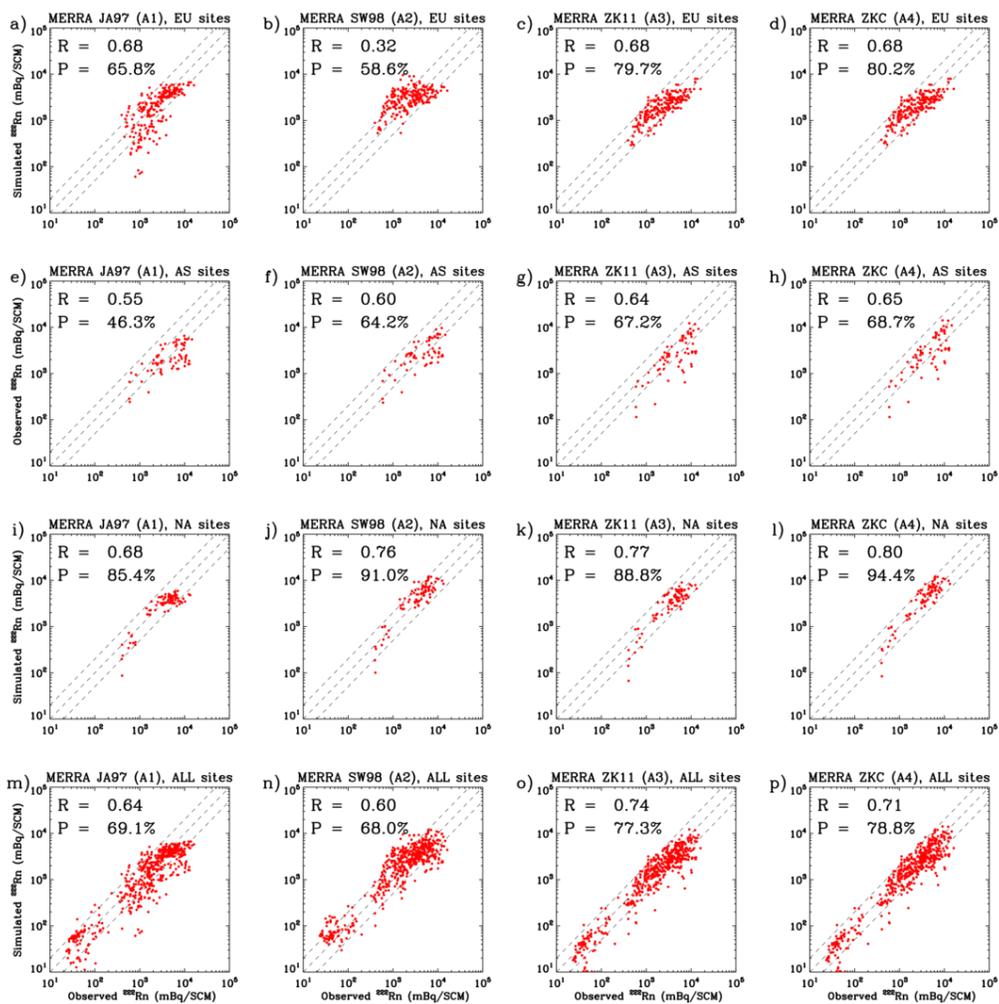
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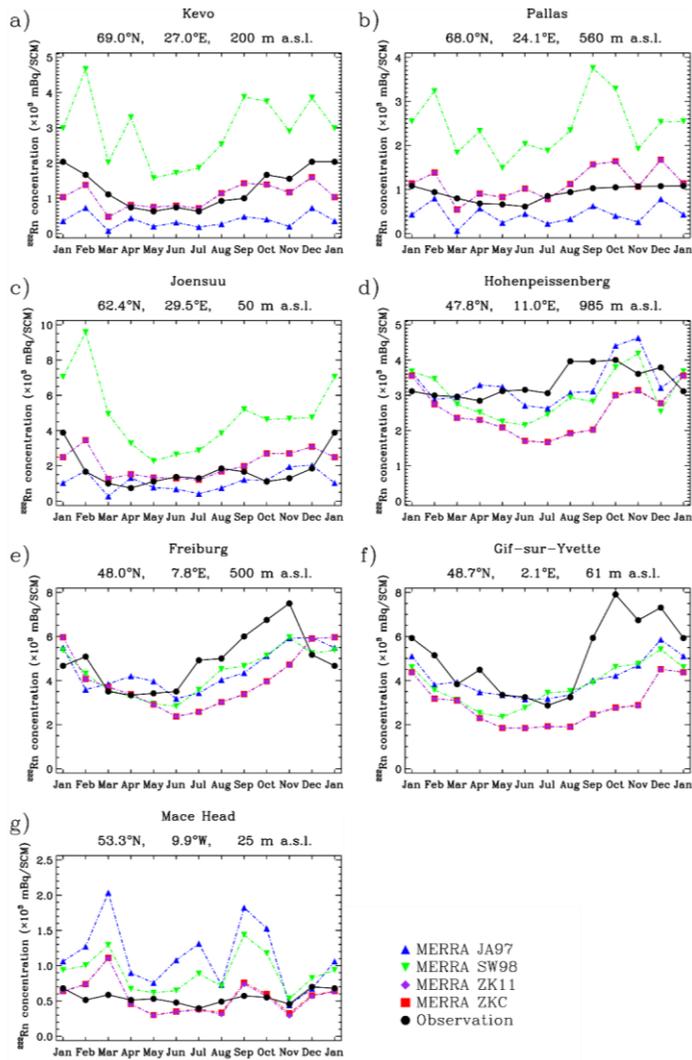


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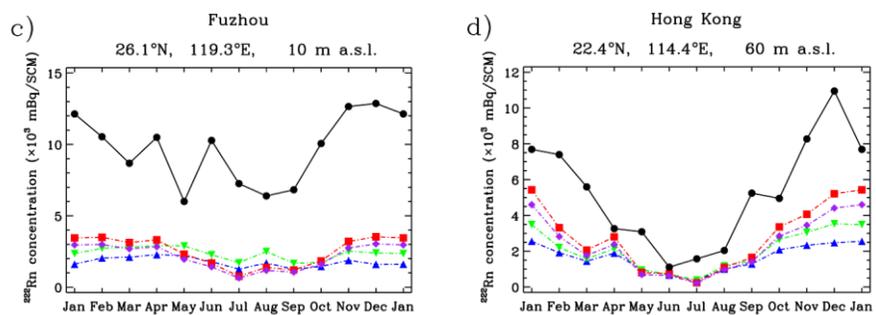
**Figure S1.** Monthly mean planetary boundary layer height (m) in MERRA for (a) January and (b) the difference between July and January 2013.



5 **Figure S2.** Same as Fig. 6, but the measured  $^{222}\text{Rn}$  concentrations at European sites are applied with scaling factors suggested by Schmithüsen et al. (2017).



**Figure S3.** Same as Fig. 8, but the measured  $^{222}\text{Rn}$  concentrations at European sites are applied with scaling factors suggested by Schmithüsen et al. (2017).



**Figure S4.** Same as Fig. 9-c and 9d, but the simulated  $^{222}\text{Rn}$  concentrations are sampled from the grid boxes where the stations are located corresponding to the site locations.