Evaluation of transport processes in MERRA/GEOS-FP driven chemical transport models using ^{222}Rn

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Introduction

This work uses an atmospheric radionuclide, ^{222}Rn, to evaluate synoptic and convective transport in chemical transport models (GEOS-Chem & GMI CTMs) driven by two meteorology data sets (MERRA & GEOS-FP).

Background

- Convective transport plays a major role in determining the transport and distribution of trace gases and aerosols in the troposphere. Convection is a sub-grid process that needs to be parameterized in global CTMs, and therefore is a source of model uncertainties.
- ^{222}Rn: A noble gas emitted from continental crust; radioactive lifetime (3.82 days) comparable to synoptic and convective transport timescales, thus an effective tracer for these transport processes.
- An accurate ^{222}Rn emission inventory is required for such model application. The current Rn emissions in GEOS-Chem and GMI are lack of spatial variability, and can be updated according to recently available inventories.
- The differing convection characteristics in NASA GMAO's MERRA and GEOS-FP assimilated meteorology data sets may result in different roles of convection in determining the distribution of trace gases and aerosols in MERRA/GEOS-FP driven CTMs.

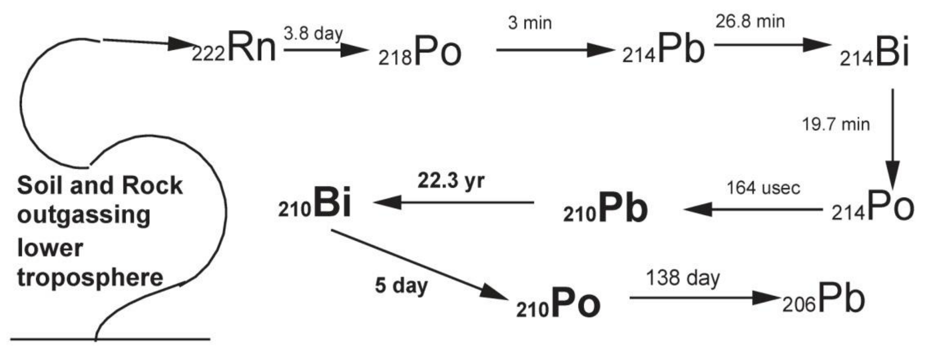

Figure 1. The sources, radioactive lifetimes, and decay schemes of tracers ^{222}Rn and its progeny. [Gaffney et al., 2004]

Goals

- Compare the global total magnitude, spatial variation and seasonality of the standard ^{222}Rn emission in GEOS-Chem with the two more recently developed emission inventories.
- Evaluate GEOS-Chem and GMI ^{222}Rn simulations with the three available ^{222}Rn emission inventories against observations at global surface sites and profile measurements, and recommend an optimal ^{222}Rn emission inventory.
- Assess and compare the characteristics of convective transport in MERRA and GEOS-FP using ^{222}Rn.

Models and Data

Models

GEOS-Chem

- v9-02 driven by MERRA and GEOS-FP.
- 2°×2.5° horizontal resolution and 47 vertical levels.
- Rn-Pb-Be simulations for a single year, 2013, when both MERRA and GEOS-FP are available.

GMI

- Global Modeling Initiative (GMI) driven by MERRA.
- 2°×2.5° horizontal resolution and 47 vertical levels.
- Rn-Pb-Be simulations for a 36-year climatology (1979-2014).

Meteorological fields

- **MERRA:** Modern-Era Retrospective Analysis for Research and Applications, a 30 year re-analysis with GEOS-5.2.0.
- **GEOS-FP:** the current version of the GMAO data assimilation system (DAS) product.

The two meteorology data sets differ in the strength of convection as shown in ***Figure 2***.

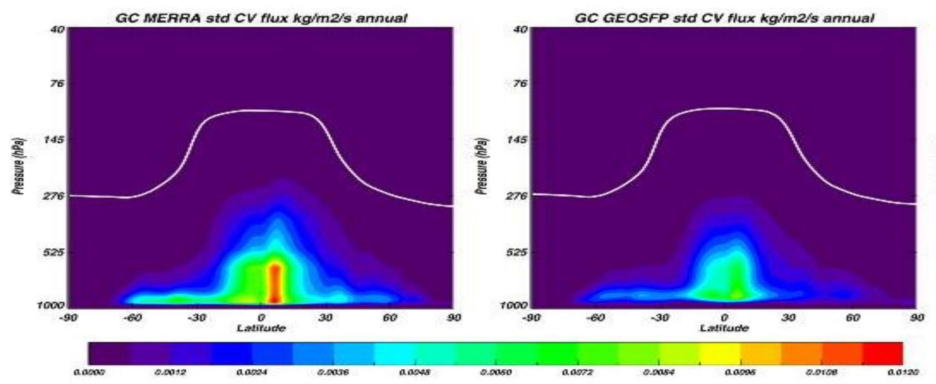

Figure 2. Annual zonal mean of convective air mass fluxes in MERRA and GEOS-FP. The white line in each figure shows averaged level of the zonal tropopause.

^{222}Rn emissions

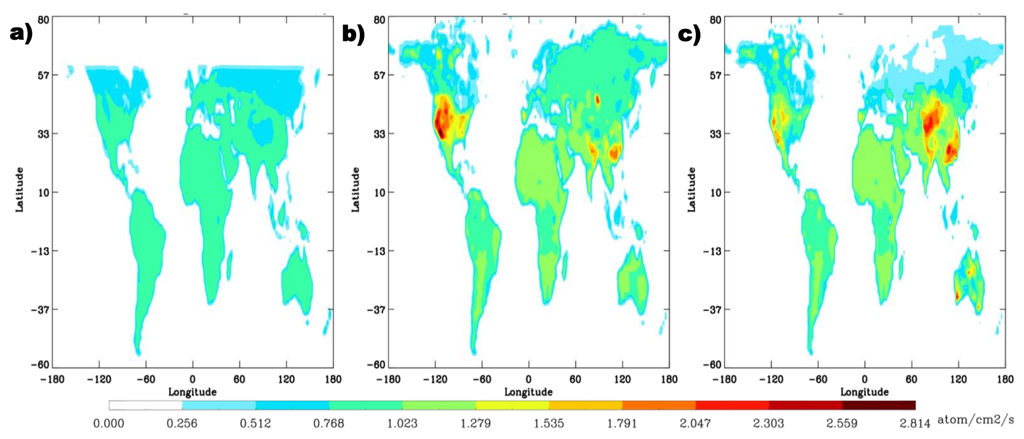

Figure 3. Global annual mean of ^{222}Rn emissions in a) GEOS-Chem Standard, b) SW1998 and c) ZK2011 inventories.

- **Standard:** (*Figure 3a*)
  - Defined in Jacob et al. (1990) .
  - Fluxes of 1.0 atoms $cm^{-2}s^{-1}$ over land and a reducing factor of 3 under freezing conditions.
  - No emissions over lakes/oceans or above 70°N/ 70°S.
- **SW1998:** (*Figure 3b*)
  - Developed by Schery and Wasiolek (1998) .
  - Based on porous media transport theory and calibration by experimental ^{222}Rn flux density data.
  - A reducing factor of 1.6 is applied for better agreement on total global emission.
- **ZK2011:** (*Figure 3c*)
  - Zhang et al. (2011) compiled recent ^{222}Rn flux measurements over Europe, North American, and Aisa, with the scaled SW1998.

Conclusions

- Compared to the relatively uniform ^{222}Rn emissions in the standard GEOS-Chem model, the Schery and Wasiolek (1998) and Zhang et al. (2011) ^{222}Rn emission inventories feature large regional variability. Using the latter inventories improves the simulated spatial variability and seasonality of ^{222}Rn in surface air.
- Among the three inventories we tested, the Zhang et al. (2011) emission inventory provides the best estimate. However, replacing North American ^{222}Rn emissions in Zhang et al. (2011) with the Schery and Wasiolek (1998) inventory would constitute an optimal global ^{222}Rn emission inventory.
- Using the Zhang et al. (2011) emission inventory, in which Asian ^{222}Rn emissions are significantly enhanced, improves the simulations but still underestimate surface ^{222}Rn in the region, supporting excessive ^{222}Rn emissions over Asia.
- GEOS-Chem ^{222}Rn simulations driven by MERRA and GEOS-FP indicate that GEOS-FP has much weaker convective transport with compensating stronger large-scale vertical advection.

Results and Discussion

Experiments

Table 1. ^{222}Rn experiments designed to evaluate ^{222}Rn emissions and convective transport in MERRA and GEOS-FP.

Exp	CTM	Rn emissions	Meteorology	Convection
A	**GEOS-Chem**	**Std**	**MERRA**	**On**
B	**GEOS-Chem**	**SW1998**	**MERRA**	**On**
C	**GEOS-Chem**	**ZK2011**	**MERRA**	**On**
D	**GEOS-Chem**	**Std**	**GEOS-FP**	**On**
E	**GEOS-Chem**	**Std**	**MERRA**	**Off**
F	**GEOS-Chem**	**Std**	**GEOS-FP**	**Off**
E	**GMI**	**Std**	**MERRA**	**On**

Seven experiments were set up as detailed in ***Table 1***.

- Exp. A-C: assess ^{222}Rn emission options.
- Exp. A, D, E and F: study the contribution of convective transport in MERRA and GEOS-FP.
- Exp. E is a long-term GMI climatology to study convection in MERRA and inter-annual variability of simulated Rn compared with observations.

^{222}Rn measurements

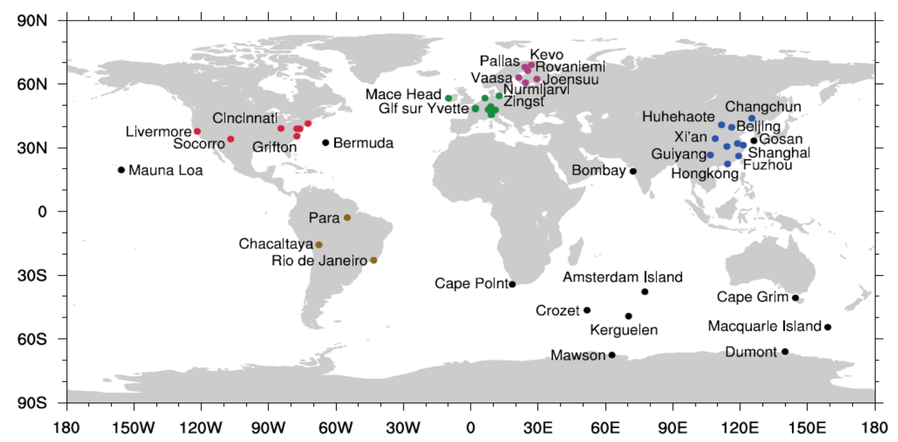

Figure 3. Locations of the surfaces sites where ^{222}Rn measurements were taken. Figure is taken from Zhang et al. (2011).

We use the surface radon observational data sets (1950s-2010s) compiled by Zhang et al. (2011).

Simulated surface ^{222}Rn concentrations

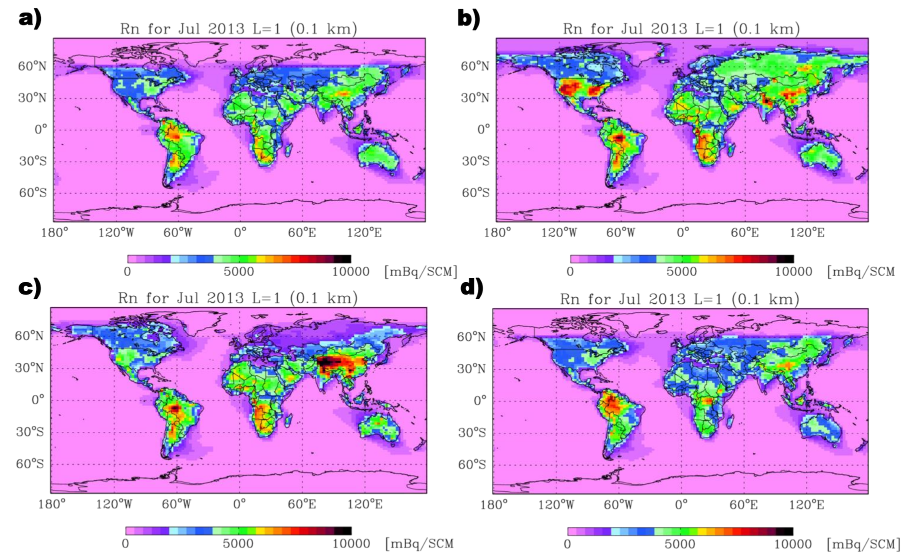

Figure 4. Simulated surface ^{222}Rn concentrations for July in experiments A-D.

- Simulated global surface ^{222}Rn distributions generally reflect the spatial variability of emissions, owing to the short lifetime of ^{222}Rn.
- Due to weaker convective transport in GEOS-FP, ^{222}Rn concentrations in tropical land regions are higher in GEOS-FP / Std (***Figure 4d***) than MERRA / Std (***Figure 4a***).

Future work

- Generate an optimized ^{222}Rn emission inventory by scaling up the emission over Asia in the ZK2011 inventory and adapting emission over NA in SW1998.
- Evaluate the deposition fluxes of ^{210}Pb (a decay daughter of ^{222}Rn) against observations, especially over Asia.

Evaluation of ^{222}Rn emissions against surface measurements

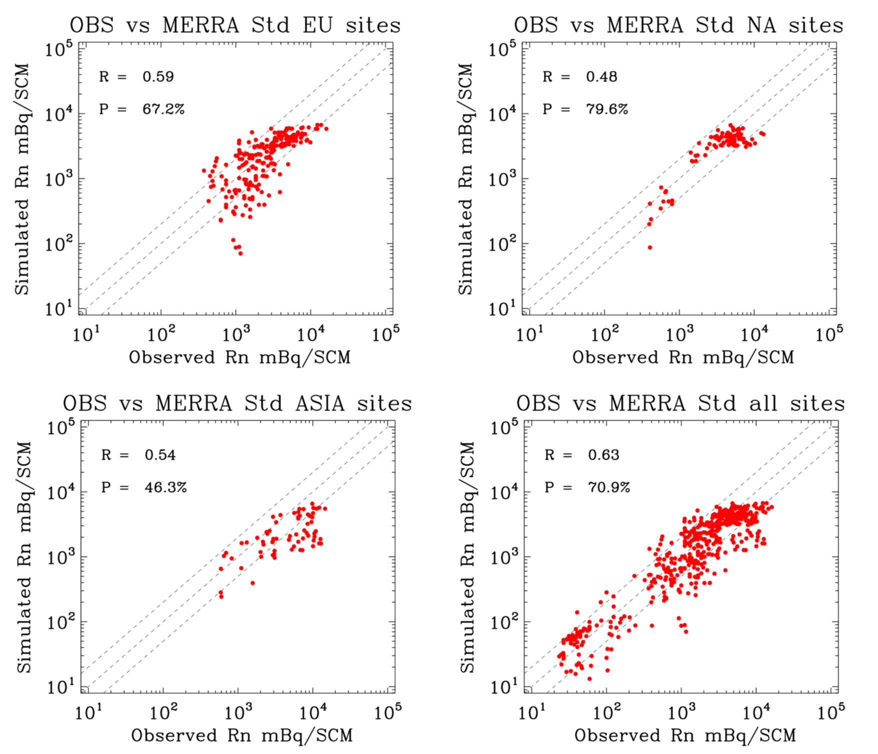

Figure 5. Evaluations of simulated surface ^{222}Rn against regional surface observations. R: correlation coefficient. Dashed lines: the range within a factor of two of the measurements. P: the percentage of the data in this range.

Table 2. Summary of evaluations against regional surface observations for each experiments using different meteorology and ^{222}Rn emissions.

Exp.	EU	NA	Asia	Globe
A	67.2%	79.6%	46.3%	70.9%
B	59.0%	81.6%	62.7%	67.8%
C	77.3%	65.3%	65.7%	73.9%
D	69.0%	73.5%	46.3%	69.6%

- All experiments show underestimated Rn concentrations over Asia (only Exp. A is shown in ***Figure. 5*** as an example).
- SW1998 has a better estimate of ^{222}Rn emissions over North America with the limitation of having only a few observation sites along the west coast.
- The experiments with ZK2011 perform better in Europe, Asia and the globe overall.

Seasonality simulated by GMI

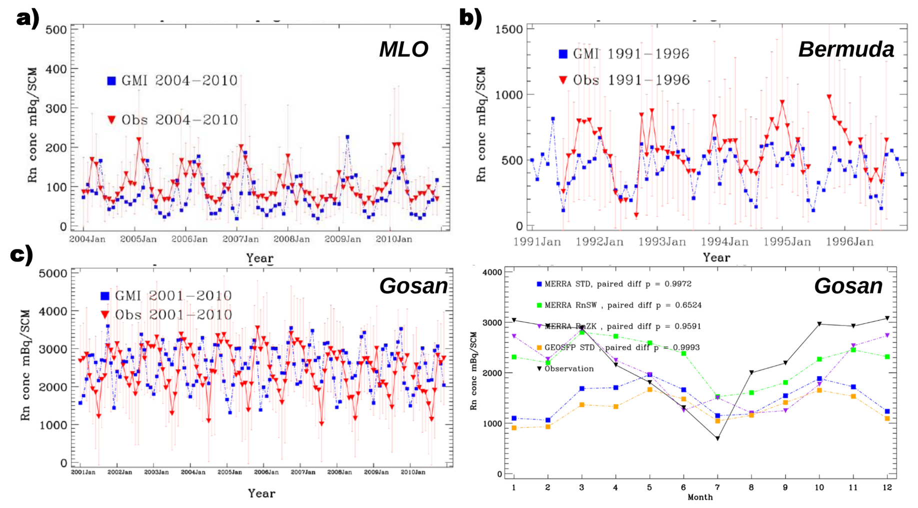

Figure 6. Observed time series of ^{222}Rn compared with GMI simulated concentrations at three sites: a) MLO, b) Bermuda, and c) Gosan and GEOS-Chem simulated concentrations at d) Gosan.

- GMI performs better at remote locations (MLO and Bermuda) because ^{222}Rn concentrations are mainly determined by long-range transport.
- GMI simulated ^{222}Rn at Gosan show a shifted phase (minimum in winter) compared to the observations (minimum in summer), which is due to inaccurate seasonality of ^{222}Rn emissions.
- Using SW1998 and ZK2011 in GEOS-Chem improve the ^{222}Rn simulation in winter at Gason.

Evaluation against measured ^{222}Rn profiles

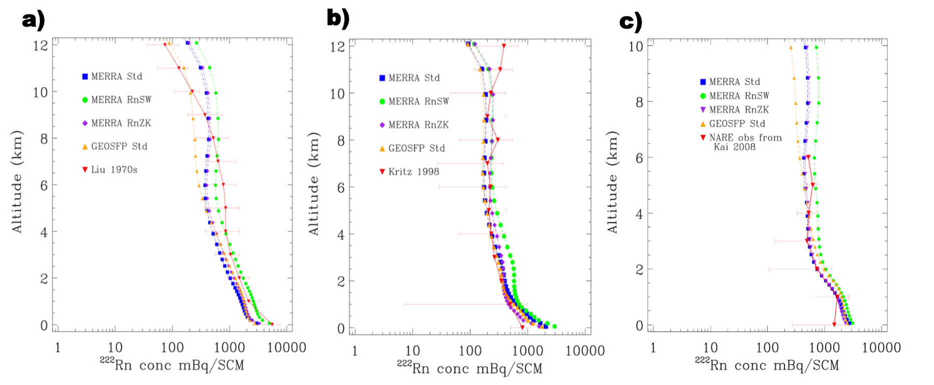

Figure 7. Comparison of simulated ^{222}Rn profiles in Experiments A-D with profiles published by Liu et al. (1984), Kritz et al. (1998) and Zaucker et al. (1996)

- Model preforms better for Kritz (U.S. western coast) and Zaucker (NA eastern seaboard) profiles than for the Liu profile, which was compiled from multiple continental sites.
- Comparison with Liu profile suggests the convection in MERRA may be too deep into the upper troposphere.
- SW1998 emission is generally higher and results in higher Rn concentrations at all altitudes.

Characteristics of convective transport in MERRA and GEOS-FP

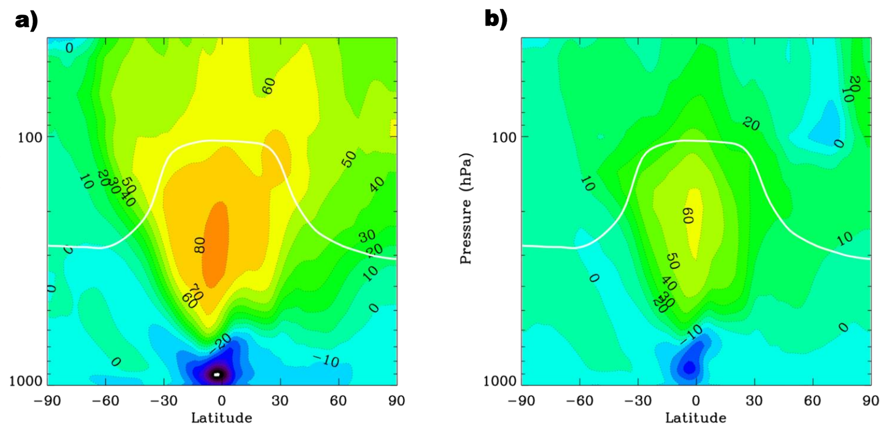

Figure 8. Percentages of annual zonal mean ^{222}Rn contributed by convective transport in (a). GEOS-Chem/MERRA, (b). GEOS-Chem/GEOS-FP, and (c). GMI/MERRA.

- Convective transport has an effect of pumping ^{222}Rn from the surface to MT/UT.
- Convective transport in G-C/MERRA contributes ~80% of ^{222}Rn in the tropical MT compared with only ~60% in G-C/GEOS-FP.

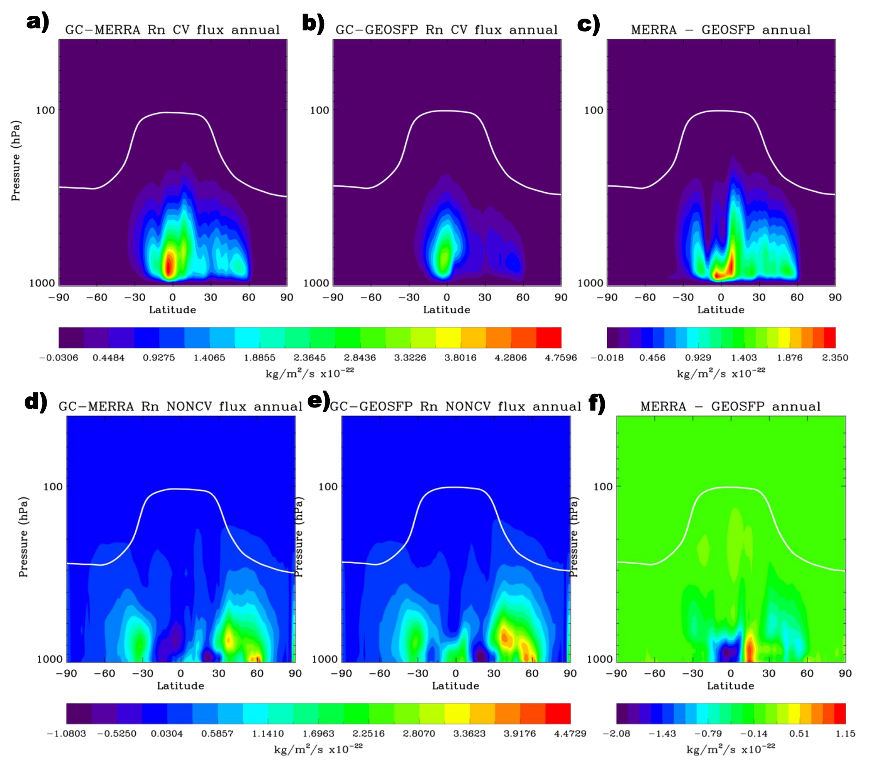

Figure 9. Upper panel: July zonal mean convective fluxes of ^{222}Rn in G-C/MERRA (a), GEOS-FP(b) and the difference (c, MERRA - GEOS-FP). Lower panel: Large scale upward transport of ^{222}Rn in GEOS-Chem driven by MERRA(d), GEOS-FP(e) and the difference (f, MERRA - GEOS-FP).

- ^{222}Rn convective fluxes in the LT in G-C/MERRA are about a factor of 1.5 larger than in G-C/GEOS-FP at northern mid-latitudes in July.
- Large-scale vertical advection fluxes of ^{222}Rn in G-C/GEOS-FP are a factor of 1.5 larger than in G-C/MERRA and compensate with the smaller convective fluxes in G-C/GEOS-FP.

References

Jacob, D. J., & Prather, M. J. (1990). Radon-222 as a test of convective transport in a general circulation model. *Tellus B*, *42*(1), 118-134.
Schery, S. D., & Wasiolek, M. A. (1998). Modeling radon flux from the earth's surface. In *Radon and Thoron in the human environment*.
Zhang, K., Feichter, J., Kazil, J., Wan, H., Zhuo, W., Griffiths, A. D., ... & Brunke, E. G. (2011). Radon activity in the lower troposphere and its impact on ionization rate: a global estimate using different radon emissions. *Atmospheric Chemistry and Physics*, *11*(15), 7817-7838.
Liu, S. C., McAfee, J. R., & Cicerone, R. J. (1984). Radon 222 and tropospheric vertical transport. *Journal of Geophysical Research: Atmospheres (1984–2012)*,*89*(D5), 7291-7297.
Kritz, M. A., Rosner, S. W., & Stockwell, D. Z. (1998). Validation of an off-line three-dimensional chemical transport model using observed radon profiles: 1. Observations. *Journal of Geophysical Research: Atmospheres (1984–2012)*,*103*(D7), 8425-8432.
Zaucker, F., Daum, P. H., Wetterauer, U., Berkowitz, C., Kromer, B., & Broecker, W. S. (1996). Atmospheric 222Rn measurements during the 1993 NARE Intensive. Journal of Geophysical Research: Atmospheres (1984–2012),101(D22), 29149-29164.

Acknowledgement

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