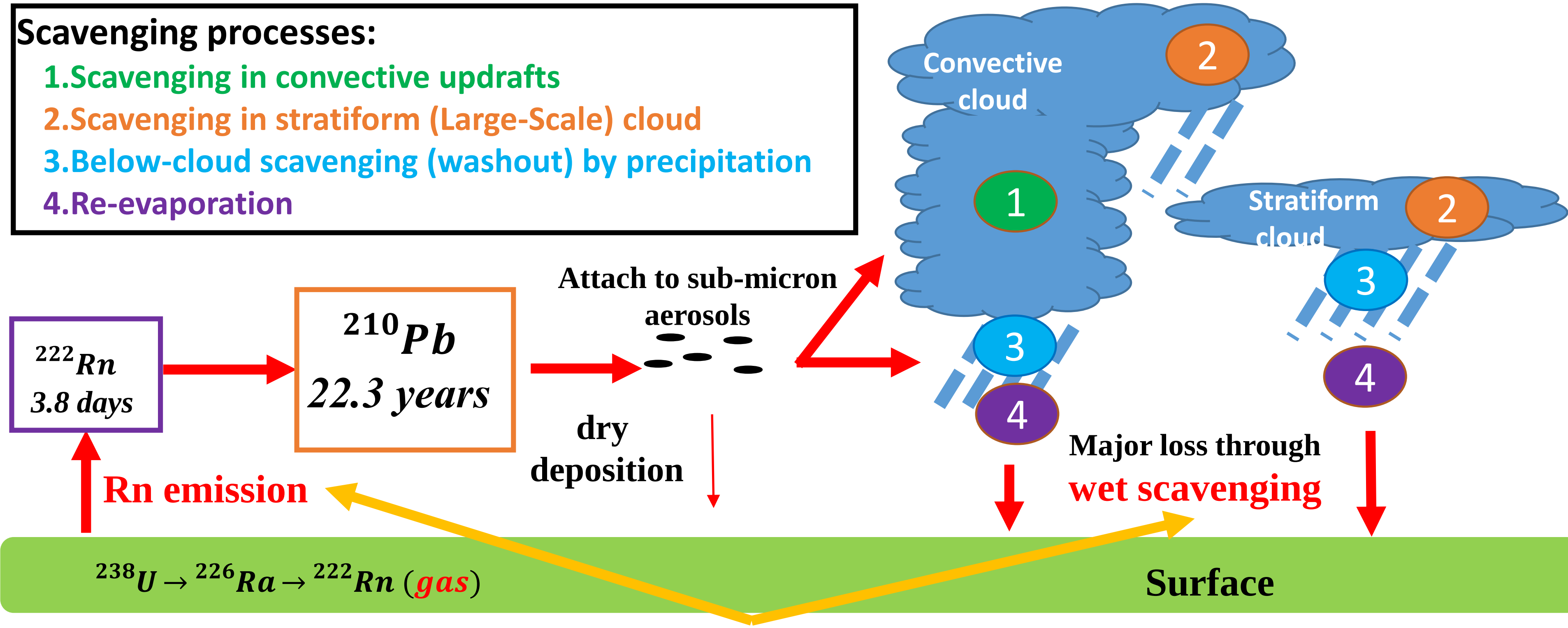
Constraints From Airborne ^{210}Pb Observations on Aerosol Scavenging and Lifetime in GEOS-Chem

Bo Zhang[1], Hongyu Liu[1], James H. Crawford[2], T. Duncan Fairlie[2], Gao Chen[2], Jack E. Dibb[3], Viral Shah[4], Daniel J. Jacob[5], Melissa P. Sulprizio[5], and Robert M. Yantosca[5]

[1] National Institute of Aerospace, Hampton, VA
[2] NASA Langley Research Center, Hampton, VA
[3] University of New Hampshire, Durham, NH
[4] University of Washington, Seattle, WA
[5] Harvard University, Cambridge, MA

National Institute of Aerospace

^{222}Rn-^{210}Pb simulation: sources and sinks

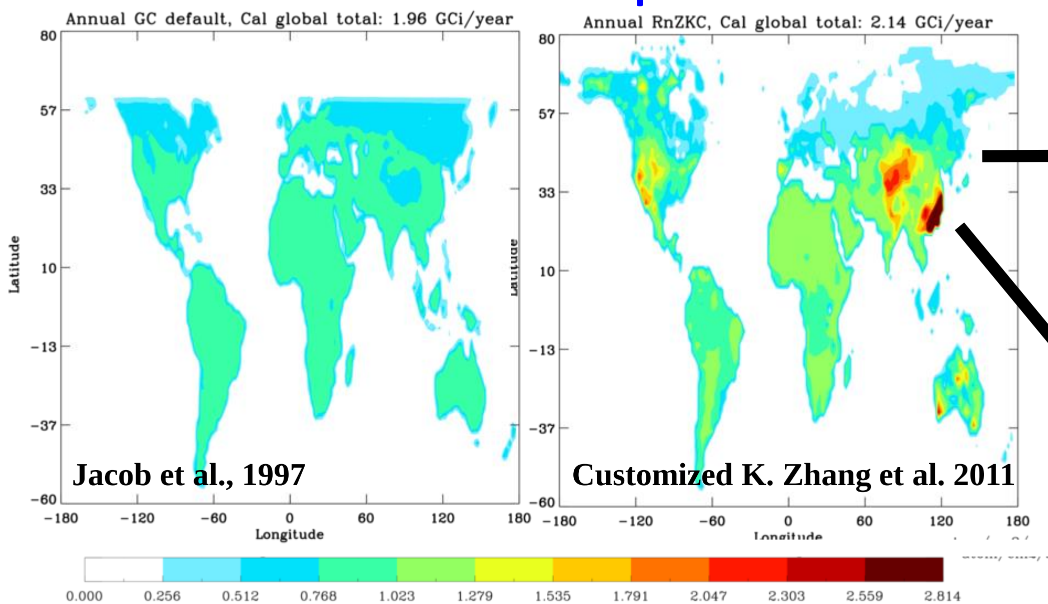


Focus of this work

- Does the updated ^{222}Rn emission improve simulated ^{222}Rn and ^{210}Pb?
- What is the observation-constrained lifetime of ^{210}Pb aerosols? How does it compare to previous work [H. Liu et al., JGR 2001; Q. Wang et al., ACP 2014]?
- What aspects of the scavenging parameterization need improving?

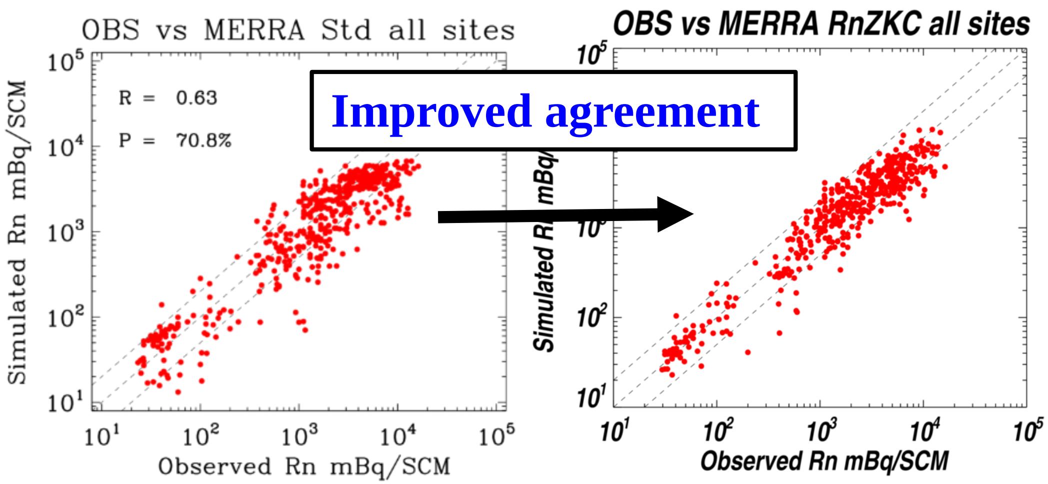
^{222}Rn emission update and evaluation

Standard emission | **Updated emission**



- Incorporated recent ^{222}Rn flux measurements in Europe, N. America, China, and Australia.
- We increased ^{222}Rn fluxes over the SE China based on comparisons with ^{222}Rn concentration and ^{210}Pb dep flux.

Comparison with worldwide surface obs



[B. Zhang, H. Liu, et al., Evaluation of transport in G-C / MERRA using ^{222}Rn, in prep. 2017.]

Wet scavenging parameterization

- In convective updrafts: $$F = 1 - exp\left(-a \cdot k \frac{\Delta Z}{\omega}\right)$$
- In stratiform clouds: $$F = a \cdot f \cdot [1 - exp(-k \cdot \Delta t)]$$
- Below cloud (washout): $$F = f[1 - exp(-k \cdot \Delta t)]$$

F : **scavenging fraction**

a : **fraction associated with cloud water, T-dependent look-up table**

k : **condensate-to-precip conversion rate; or scavenging rate constant in washout**

f : **cloud or precipitation areal fraction**

ΔZ : rising distance in updraft

ω : **updraft velocity**

Look-up table for a in 3 temperature ranges (^{210}Pb aerosol)

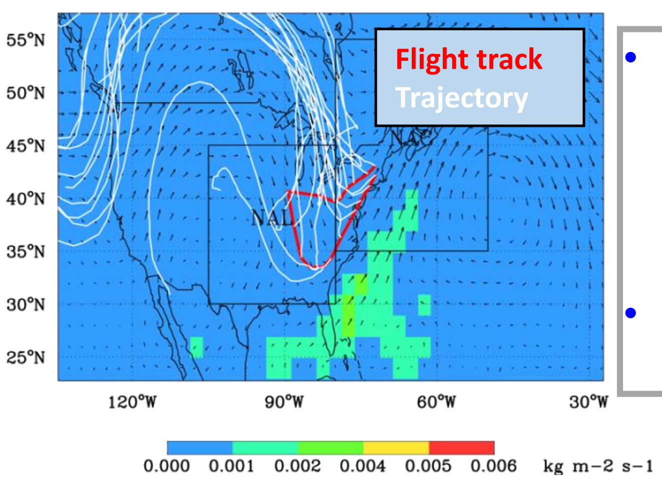
	T < 237K,	237K <= T < 258K	T > 258K
for cvrain	1.0	0.5	1.0
for lsrain	1.0	0.0	1.0

^{210}Pb measurements from NASA aircraft campaigns provide strong constraint for scavenging parameterization

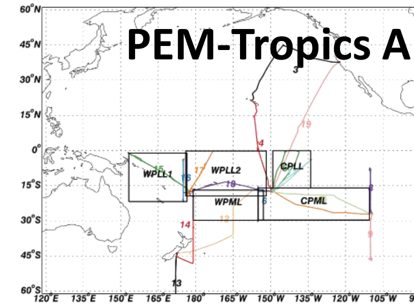
Table 1. List of NASA field campaigns where aircraft ^{210}Pb and ^{7}Be measurements were made by Jack Dibb (collaborator) and his colleagues at University of New Hampshire.

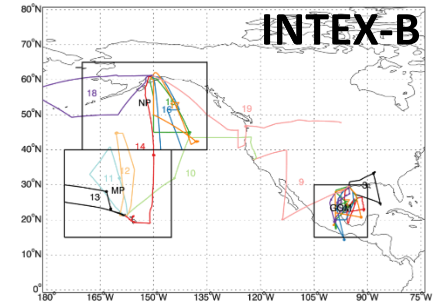
Campaign	Region	Period
PEM West A*	NW Pacific	September-October 1991
PEM West B*	NW Pacific	February-March 1994
PEM Tropics A*	southern tropical regions of the Pacific Ocean	August -September 1996
SUCCESS*	Atlantic Ocean, Gulf of Mexico, & Eastern USA	April - May 1996
SONEX*	North Atlantic Flight Corridor	October-November 1997
PEM Tropics B	tropical Pacific	March-April 1999
TRACE P	NW Pacific	March-April 2001
TOPSE	North America	February - May 2000
INTEX NA	North America	summer 2004
INTEX B	North America	spring 2006
TC4	Eastern Pacific Ocean	July - August 2007

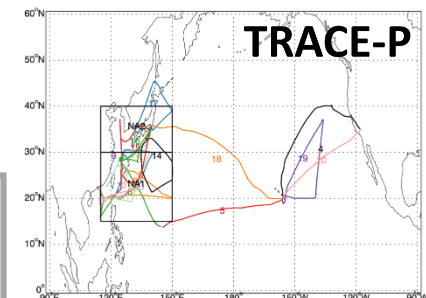
*These campaigns also include ^{10}Be

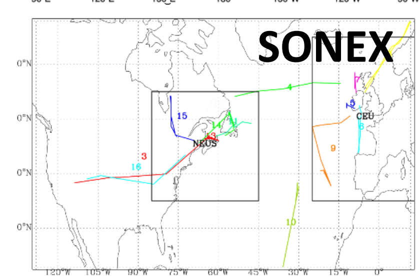


- **Calculated 5-day backward "trajectories" and sampled convective snow, convective rain, LS snow, and LS rain along the "trajectories".**
- **Categorize observations according to precipitation types and amount.**

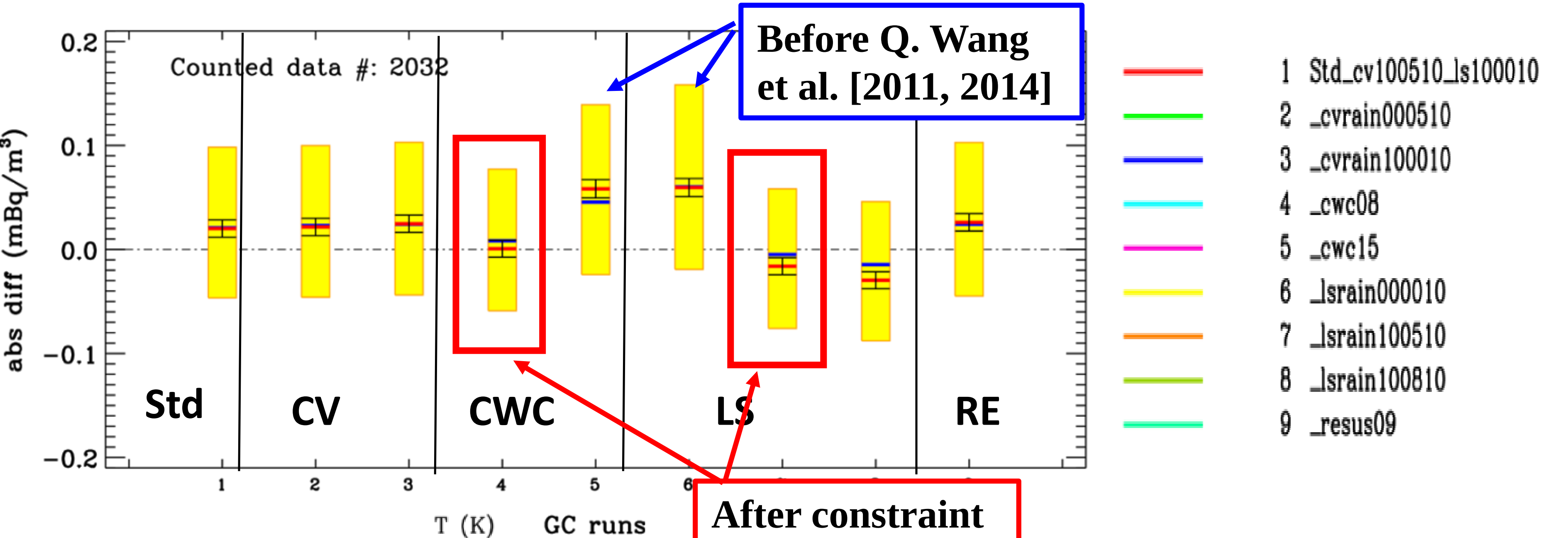


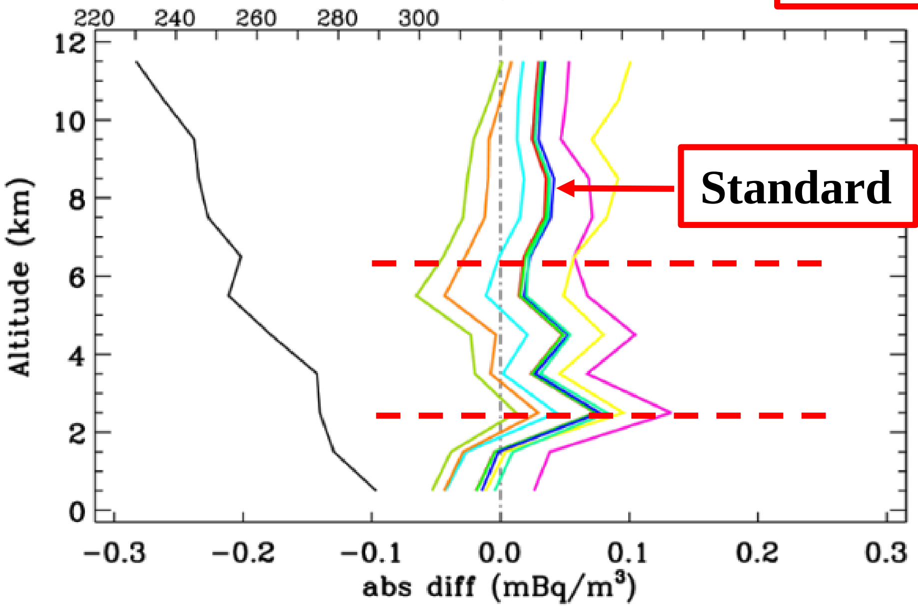






Evaluation of selected scavenging processes (v11-01f 2x2.5 MERRA)



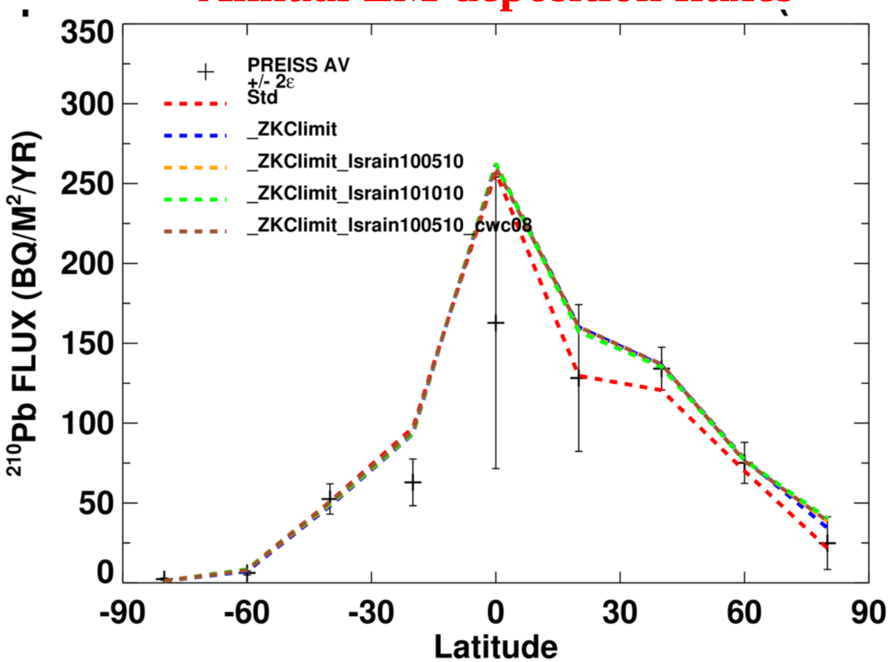


- **Model experiments suggest that scavenging in LS mixed-phase clouds (237 K < T < 258 K) should be incorporated in the model.**
- **Evidence is out there, e.g., riming in mixed-phase clouds (L. Qi et al., ACP 2017).**

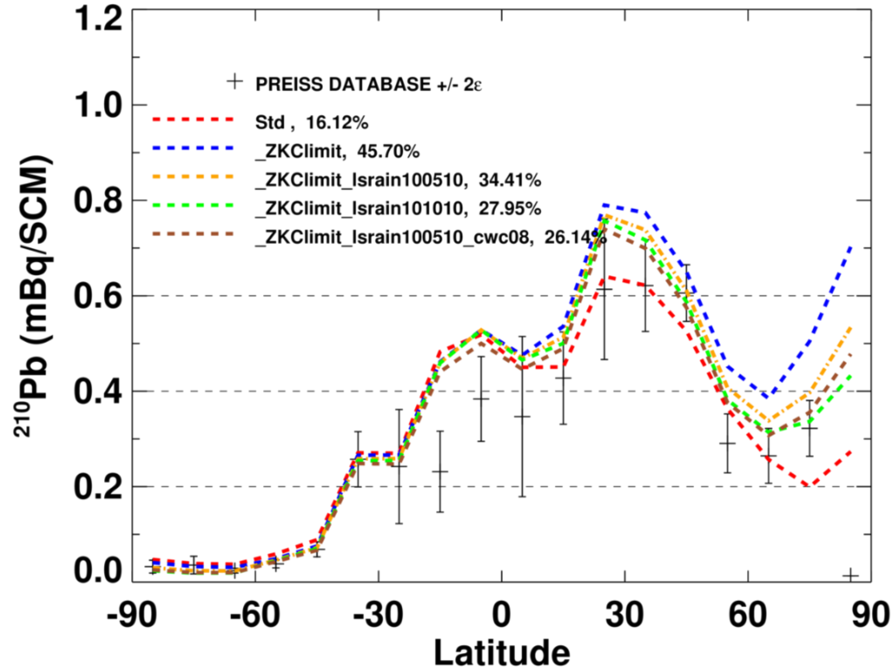
Evaluation w/ observed ^{210}Pb dep. fluxes and surface conc.

PREISS database: Measurements of surface ^{210}Pb conc. during 1960-1990.

Annual ZM deposition fluxes

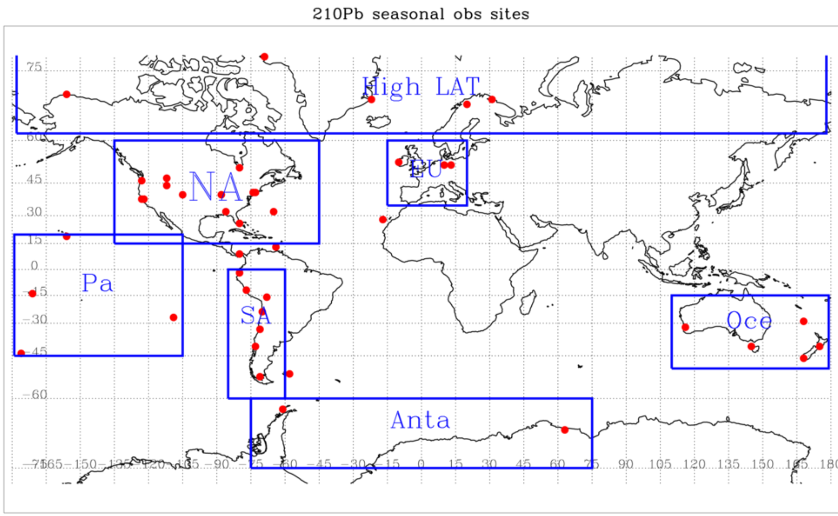


Annual ZM surface conc.



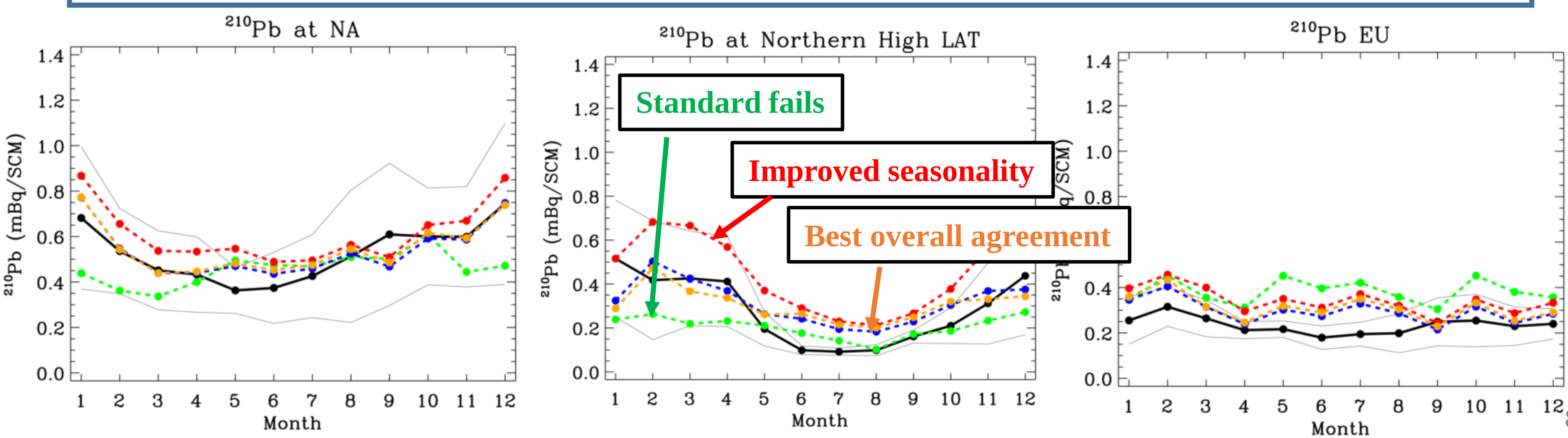
- The updated emission resulted in higher deposition fluxes in the NH, but still reasonably consistent with observations.
- Model w/ the updated emission overestimates surface concentrations, suggesting stronger scavenging is required in the model.
- Increasing scavenging in large-scale precipitation improved simulated surface concentrations.

Evaluation w/ observed ^{210}Pb seasonality



- **Updated emission resulted in improved seasonality over NA and at high-latitude sites.**
- **Improved surface ^{210}Pb concentrations with increased LS in-cloud scavenging.**

- **Observations**
- **Standard**
- **Updated Rn emission**
- **Updated Rn emission + LS mixed-phase scav**
- **Updated Rn emission + LS mixed-phase scav + reduced LWC**



Simulated tropospheric ^{210}Pb lifetime

$$lifetime = \frac{burden}{"wet + dry"\ deposition}$$

Lifetime (days)	Global Troposphere	60N - 90N	30N - 60N	15S - 15N
Standard	8.5	13.8	8.1	5.6
Emission update	8.3	12.3	8.1	5.5
lsrain100510 + cwc0.8	6.9	8.1	6.3	5.0
lsrain101010	7.0	7.7	6.4	5.1

- **Global tropospheric ^{210}Pb lifetime reduced from 8.5 to ~ 6.9 days with LS mixed-phase cloud scavenging.**
- **Lifetime in northern high-latitude regions reduced ~ 42% (from 13.8 to 8.1 days), which contributed to the most change in global lifetime.**

Summary and Conclusions

- **Updated ^{222}Rn emission improves model simulations of both ^{222}Rn (conc. & seasonality) and ^{210}Pb (seasonality).**

- **While the previously implemented ice/snow scavenging improves ^{210}Pb simulation, our evaluation with aircraft /surface observations supports the incorporation of scavenging in mixed-phase clouds.**

- **Our best estimated global ^{210}Pb lifetime is ~6.9 days, which is ~ 2 days shorter than previously reported [H. Liu et al., JGR 2001].**

- **The optimized scavenging parameters in this work may be only suitable for the MERRA input meteorology, but the resulting observation-constrained ^{210}Pb aerosol lifetime can serve as a reference for other global models.**

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