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Variability and trends of upper-tropospheric aerosols over the Asian summer monsoon region: an AeroCom multi-model study

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S1. Transport and Removal tracers for AeroCom Phase-III model experiments

In order to diagnose the inter-model differences and the inter-annual variability of characteristics in transport and removal processes for the current AeroCom Phase III model experiments, it is recommended to implement common tracers of transport and dry/wet removal across all models for all simulated years. These requirements are described below. One-year spin-up is recommended.

S1.1 Transport tracer: CO with prescribed source and sink

We will use CO with prescribed monthly emissions for 2010 and a 50-day lifetime (exponential decay rate = 2.315×10^{-7} s^{-1}) as a common transport tracer for all models and for all simulated years. Sources of CO are from (1) direct emissions from anthropogenic and biomass burning emissions, (2) production from non-methane volatile organic compounds (NMVOC) oxidation, and (3) from methane oxidation. To simplify the tracer implementation, we treat the atmospheric production of CO from all NMVOC oxidation in a manner of direct emission, as described below.

- Prescribed sources:
  - CO from anthropogenic (anthro) emissions: CMIP6, 2010, annual total = 617.2 Tg yr^{-1}
  - CO from biomass burning (bb) emissions: CMIP6, 2010, annual total = 336.8 Tg yr^{-1}
  - CO from anthro NMVOC oxidation: Assuming it is in the order of 0.20 of the direct anthropogenic emission of CO (Bian et al., 2007). Annual total = 123.4 Tg yr^{-1}
  - CO from bb NMVOC oxidation: Assuming it is in the order of 0.11 of the direct biomass burning (bb) emission of CO (Bian et al., 2007). Annual total = 37.1 Tg yr^{-1}
  - CO from biogenic (bio) NMVOC oxidation: Assuming a CO yield of 0.2 from the oxidation of isoprene, monoterpene, and other biogenic NMVOC species (Bian et al., 2007). The CO from biogenic NMVOC is calculated based on the 2010 NMVOC emissions in the GMI model with an annual total of 254.5 Tg yr^{-1}
  - CO from CH_4 oxidation: assuming a fixed uniform CH_4 concentration of 1760 ppbv and a CH_4 decay rate = 3.73×10^{-9} s^{-1} (lifetime 8.5 years) to produce CO with a molar yield of 0.86
- Annual total source of transport tracer CO (excluding CO from CH_4 oxidation) is 1368 Tg/yr^{-1}, as listed in Table S1, and the spatial distributions of each source are displayed in Fig. S1.

Table S1. Annual sources of CO (used for TRCO50).

Source	**Direct emission**	**NMVOC oxidation**	**Total (Tg yr^{-1})**
Anthropogenic	616.6	123.3	739.9
Biomass burning	336.8	37.0	373.8
Biogenic	---	254.5	254.5
Total	953.4	414.8	**1368.2**

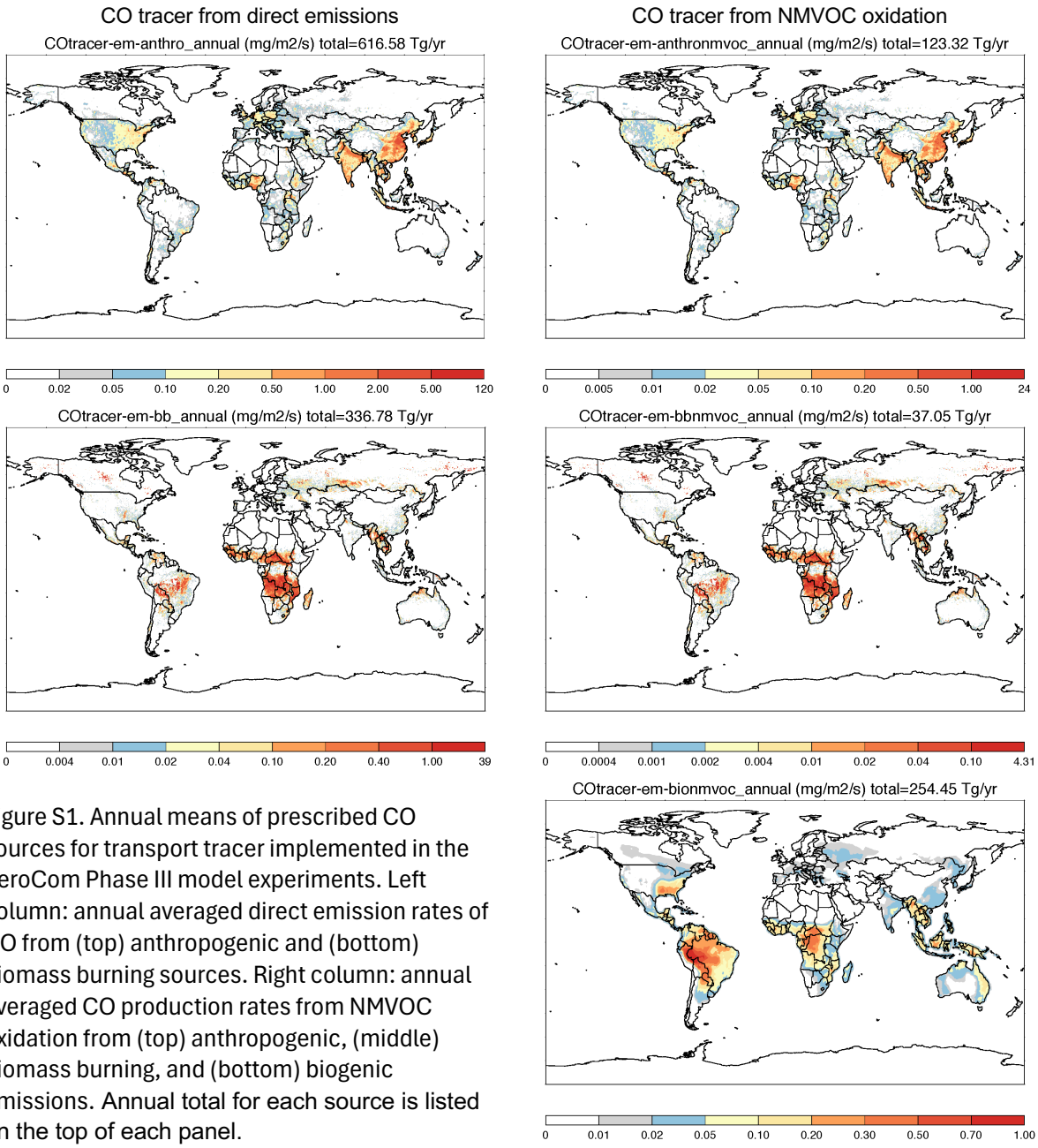

Figure S1. Annual means of prescribed CO sources for transport tracer implemented in the AeroCom Phase III model experiments. Left column: annual averaged direct emission rates of CO from (top) anthropogenic and (bottom) biomass burning sources. Right column: annual averaged CO production rates from NMVOC oxidation from (top) anthropogenic, (middle) biomass burning, and (bottom) biogenic emissions. Annual total for each source is listed on the top of each panel.

S1.2 Deposition tracer: ^{210}Pb (lead-210) with common source and prescribed sink

^{210}Pb is formed from ^{222}Rn decay and attached to fine-mode aerosols that will experience the same removal processes as soluble aerosols. Prescribed emissions of ^{222}Rn and removals of ^{222}Rn and ^{210}Pb are listed below.

- ^{222}Rn emission from land: The prescribed monthly varying ^{222}Rn emission dataset from Zhang, B. et al., 2019 (modified from Zhang K. et al., 2011) will be used for all simulations. Total annual emission is 14.2 kg yr^{-1}.
- ^{210}Pb production from the ^{222}Rn radio-active decay: A fixed first order decay rate of ^{222}Rn at 2.11x10^{-6} s^{-1} (equivalent to 5.5 days of lifetime or 3.8 days of half-life) is prescribed to produce ^{210}Pb. No other removal of ^{222}Rn.
- ^{210}Pb removal: the dry and wet deposition should be implemented in the models in the way as if it were sulfate.

Figure S2 shows the annual average ^{222}Rn emission rate and its spatial distribution.

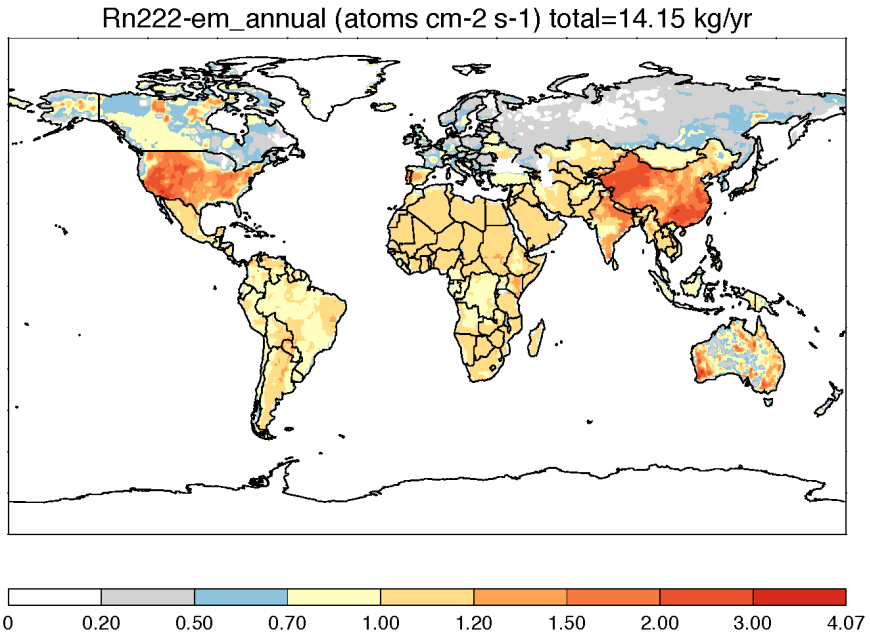

Figure S2. Annual averaged ^{222}Rn emission rate. (Note: 1 mole ^{222}Rn = 222 g = 6.023x10^{23} atoms.) Total annual emission (in kg yr^{-1}) is listed on the top of the panel.

S2. Multi-model ensemble means and inter-model spread

Table S2. Multi-model (excluding ECHAM6-HAMMOZ) ensemble means, standard deviation (Stdev), and coefficient of variation (CV in %) of AEC, TR_{CO50}, and $TR_{Pb/Rn}$ over the ASMA region at 150 hPa for each August during 2000–2018, as presented in Fig. 10 in the main body of the manuscript. (Highlighted is the August 2012 case presented in detail in the manuscript.)

Year	AEC (10^3 km^{-1})	Stdev AEC	CV %	TR_{CO50} (ppb)	Stdev TR_{CO50}	CV %	$TR_{Pb/Rn}$ (kg kg^{-1})	Stdev Pb/Rn	CV %
2000	1.022	0.821	80.33	104.40	4.93	4.72	0.803	0.296	36.86
2001	0.962	0.770	80.04	97.27	6.35	6.53	0.805	0.292	36.27
2002	1.014	0.799	78.80	97.48	2.40	2.46	0.816	0.301	36.89
2003	0.988	0.752	76.11	101.80	6.16	6.05	0.773	0.276	35.71
2004	0.951	0.741	77.92	94.79	7.29	7.69	0.789	0.285	36.12
2005	1.027	0.793	77.22	101.58	4.04	3.98	0.799	0.276	34.54
2006	1.186	0.761	64.17	97.11	3.29	3.39	0.806	0.308	38.21
2007	1.082	0.875	80.87	100.66	5.33	5.30	0.797	0.300	37.64
2008	1.267	0.960	75.77	101.56	4.67	4.60	0.804	0.286	35.57
2009	1.032	0.769	74.52	99.93	4.24	4.24	0.817	0.294	35.99
2010	1.257	1.078	85.76	98.94	4.14	4.18	0.758	0.257	33.91
2011	1.239	0.934	75.38	99.60	3.06	3.07	0.796	0.286	35.93
2012	1.219	0.964	79.08	105.20	3.46	3.29	0.792	0.275	34.72
2013	1.215	0.948	78.02	97.40	3.22	3.31	0.789	0.281	35.61
2014	1.190	0.939	78.91	99.54	3.90	3.92	0.796	0.287	36.06
2015	1.235	0.824	66.72	93.17	5.45	5.85	0.814	0.288	35.38
2016	1.233	0.968	78.51	102.21	4.68	4.58	0.793	0.270	34.05
2017	1.127	0.855	75.87	101.56	2.93	2.88	0.789	0.276	34.98
2018	1.179	0.932	79.05	102.75	4.15	4.04	0.812	0.281	34.61

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