

*Supplemental of*

# **Long-term historical trends in air pollutant emissions in Asia: Regional Emission inventory in ASia (REAS) version 3.1**

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## Supplement Figures

**Figures S1, S3, S5, S7, S9, and S11** show emissions of (a) SO<sub>2</sub>, (b) NO<sub>x</sub>, (c) CO, (d) CO<sub>2</sub>, (e) PM<sub>10</sub>, (f) PM<sub>2.5</sub>, (g) BC, (h) OC, (i) NMVOC, and (j) NH<sub>3</sub> from major sectors during 1950-2015 in China, India, Japan, Southeast Asia (SEA), East Asia other than China and Japan (OEA), and South Asia other than India (OSA), respectively. (Sectors for (a)-(h): PP = Power plants, IND = Industry, ROAD = Road transport, OTRA = Other transport, RESI = Residential, and ODOM = Other domestic; Sectors for (i): CMB = Combustion, ROAD = Road transport (including both tail pipe and evaporative emissions), INDPRC = Industrial processes, EXT = Extraction processes, PAINT = Paint use, SLV = Solvent use, and WST = Waste treatment; Sectors for (j): CMB = Combustion, MM = Manure management, FER = Fertilizer application, HUMAN = Human perspiration and respiration, and LTRN = Latrines, INDPRC = Industrial processes.)

**Figures S2, S4, S6, S8, S10, and S12** provide emissions of (a) SO<sub>2</sub>, (b) NO<sub>x</sub>, (c) CO, (d) CO<sub>2</sub>, (e) PM<sub>10</sub>, (f) PM<sub>2.5</sub>, (g) BC, (h) OC, (i) NMVOC, and (j) NH<sub>3</sub> from each fuel type during 1950-2015 in China, India, Japan, SEA, OEA, and OSA, respectively. Emissions from non-combustion sources are not included in (i) NMVOC and (j) NH<sub>3</sub> to show contributions from fuel types clearly because majority of their emissions are from non-combustion sources. In Japan, emissions from cement production were estimated not by fuel consumption, but based on production amounts of cement in each kiln type. Therefore, contributions from total emissions from cement kiln combustion are included in CEMK (see below). For brick and lime production, all emissions estimated based on production amounts are included in NCMB (see below) (Fuel types: COAL = Primary coal, DC = Secondary coal, NGAS = Natural gas, OGAS = Other gas fuels, LF = Light oil fuels, MD = Diesel oil, HF = Heavy oil fuels, BF = Biofuels, OTH = Other fuels, NCMB = Non-combustion sources, and CEMK = combustion emissions from cement kilns (only for Japan))

**Figure S13** illustrates grid maps of annual emissions of CO<sub>2</sub> and PM<sub>10</sub> in 1965 and 2015.

**Figures S14 and S15** compare CO, NMVOC, NH<sub>3</sub>, PM<sub>10</sub>, PM<sub>2.5</sub>, and OC emissions in REASv3.1 with other published estimates for China and India, respectively.

**Figures S16-S19** compare emissions of SO<sub>2</sub>, NO<sub>x</sub>, BC, CO, NMVOC, NH<sub>3</sub>, PM<sub>10</sub>, PM<sub>2.5</sub>, and OC in REASv3.1 with other published estimates for Japan, SEA, OEA, and OSA, respectively.

**Figures S20** compares relative ratios of emissions from China, India, Japan, SEA, OEA, and OSA for CO, NMVOC, NH<sub>3</sub>, PM<sub>10</sub>, PM<sub>2.5</sub>, and OC among REASv3.1, CEDS, and EDGARv4.3.2.

## China Sector

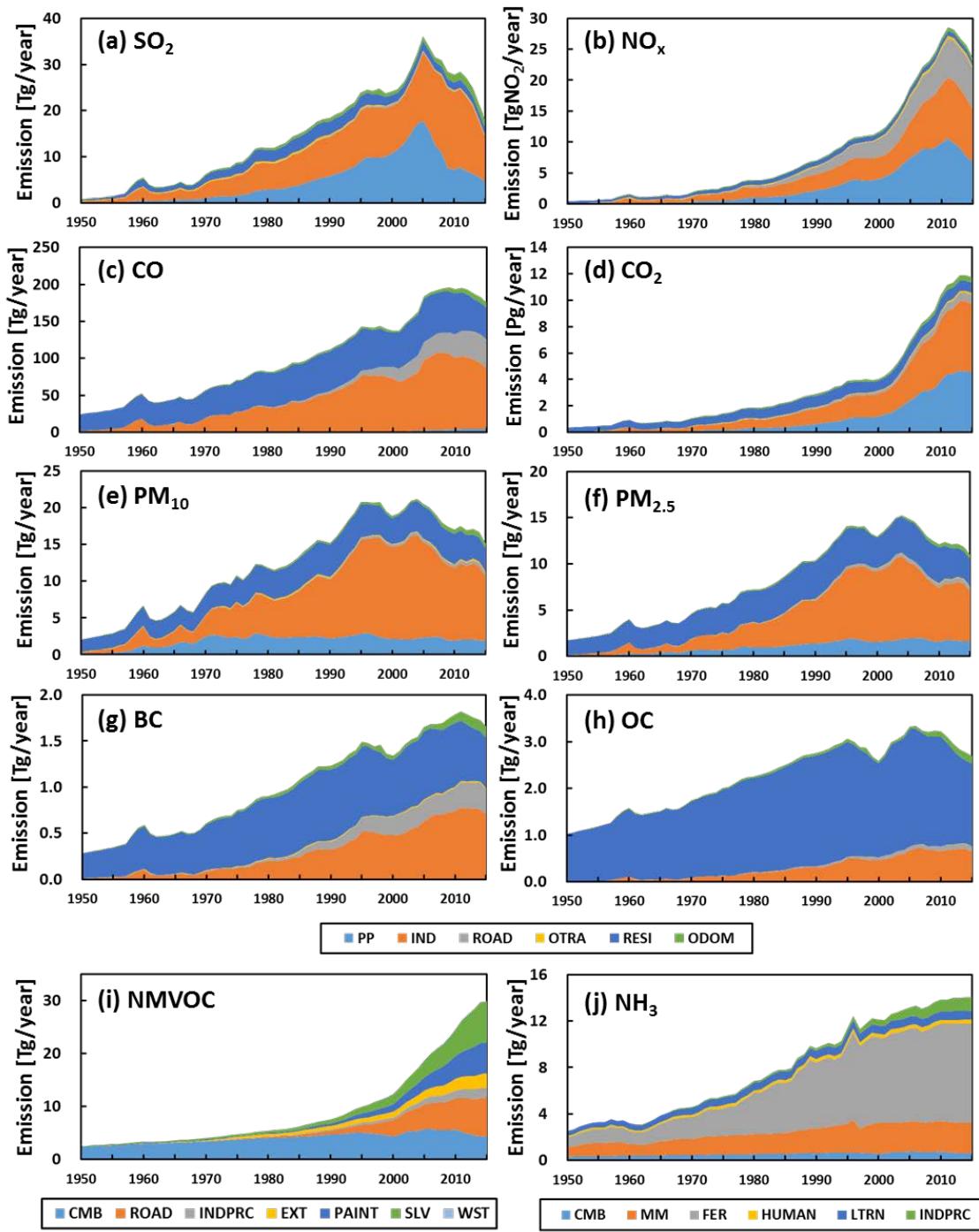


Figure S1: Emissions of (a)  $\text{SO}_2$ , (b)  $\text{NO}_x$ , (c) CO, (d)  $\text{CO}_2$ , (e)  $\text{PM}_{10}$ , (f)  $\text{PM}_{2.5}$ , (g) BC, (h) OC, (i) NMVOC, and (j)  $\text{NH}_3$  from major sectors in China from 1950 to 2015.

## China Fuel

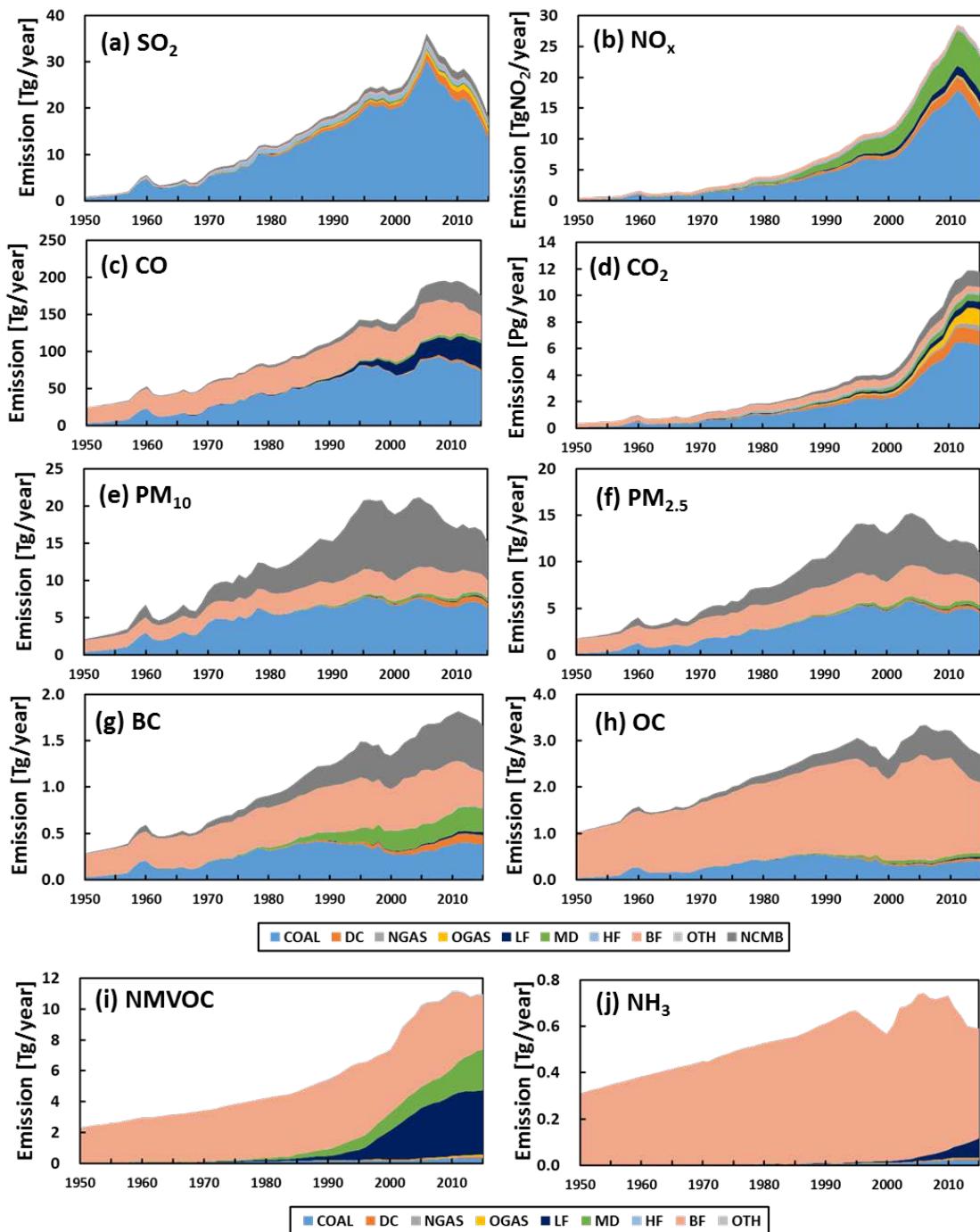


Figure S2: Emissions of (a)  $\text{SO}_2$ , (b)  $\text{NO}_x$ , (c) CO, (d)  $\text{CO}_2$ , (e)  $\text{PM}_{10}$ , (f)  $\text{PM}_{2.5}$ , (g) BC, (h) OC, (i) NMVOC, and (j)  $\text{NH}_3$  from each fuel type in China from 1950 to 2015.

## India Sector

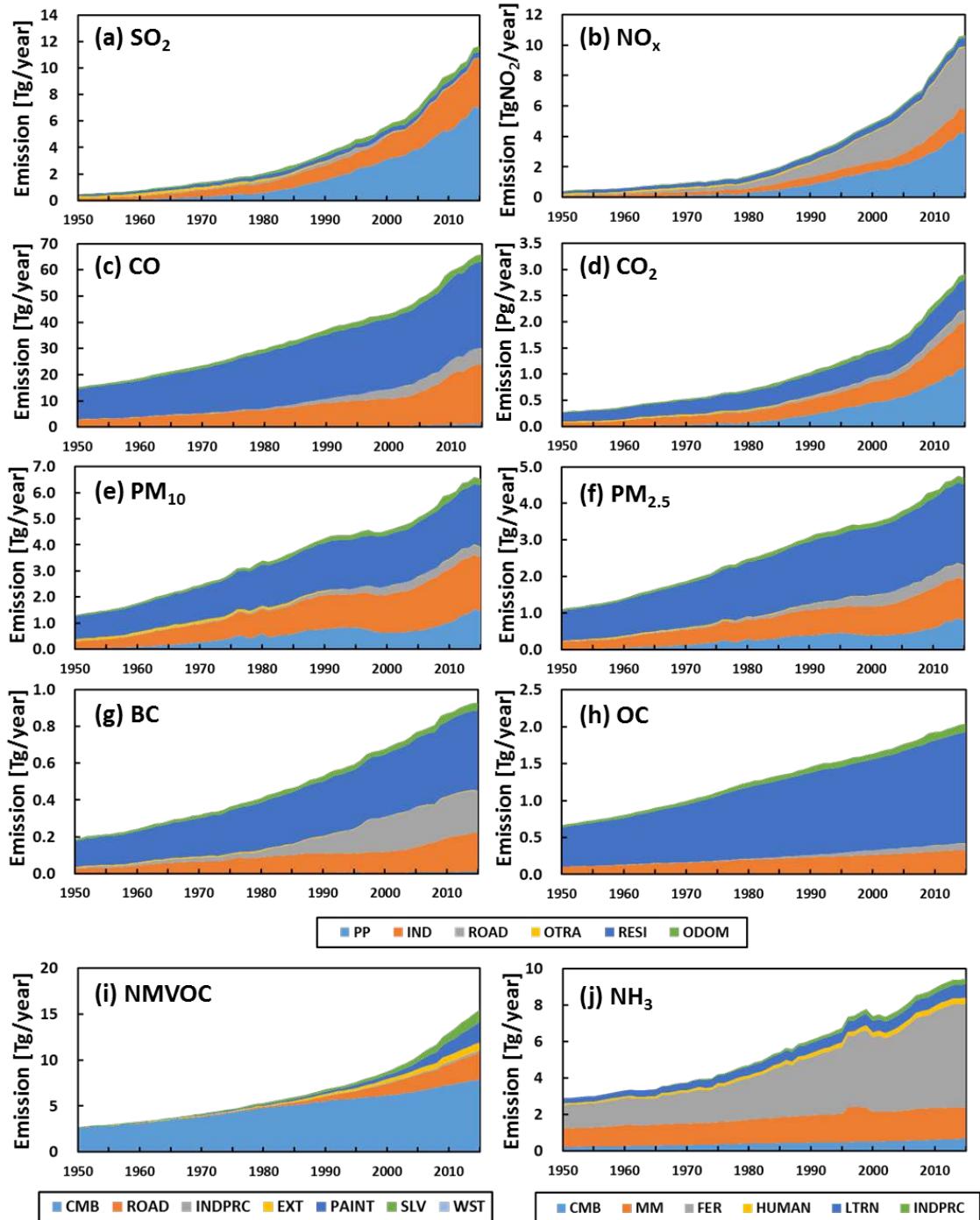
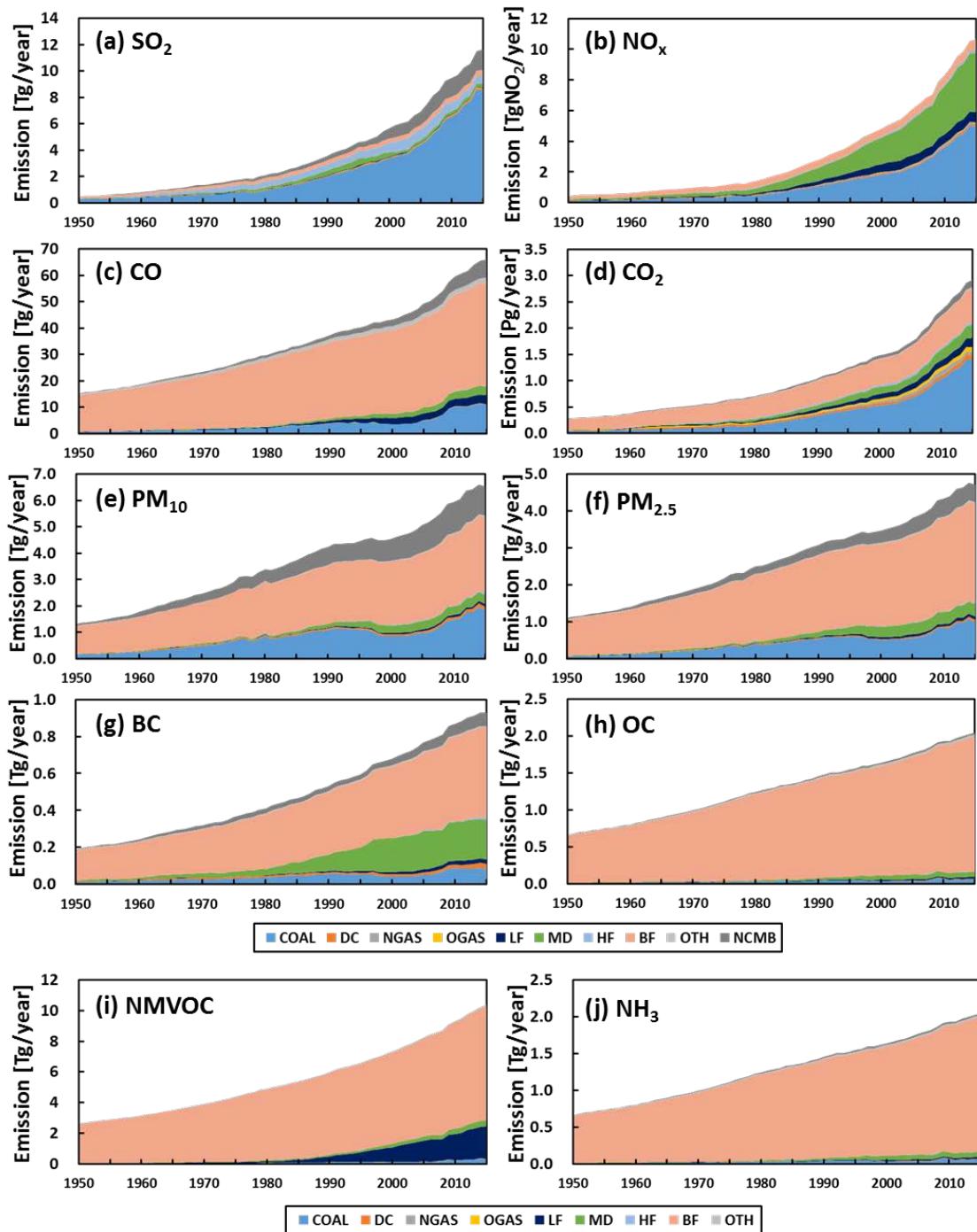


Figure S3: Emissions of (a)  $\text{SO}_2$ , (b)  $\text{NO}_x$ , (c)  $\text{CO}$ , (d)  $\text{CO}_2$ , (e)  $\text{PM}_{10}$ , (f)  $\text{PM}_{2.5}$ , (g)  $\text{BC}$ , (h)  $\text{OC}$ , (i)  $\text{NMVOC}$ , and (j)  $\text{NH}_3$  from major sectors in India from 1950 to 2015.

## India Fuel



45 Figure S4: Emissions of (a) SO<sub>2</sub>, (b) NO<sub>x</sub>, (c) CO, (d) CO<sub>2</sub>, (e) PM<sub>10</sub>, (f) PM<sub>2.5</sub>, (g) BC, (h) OC, (i) NMVOC, and (j) NH<sub>3</sub> from each fuel type in India from 1950 to 2015.

## Japan Sector

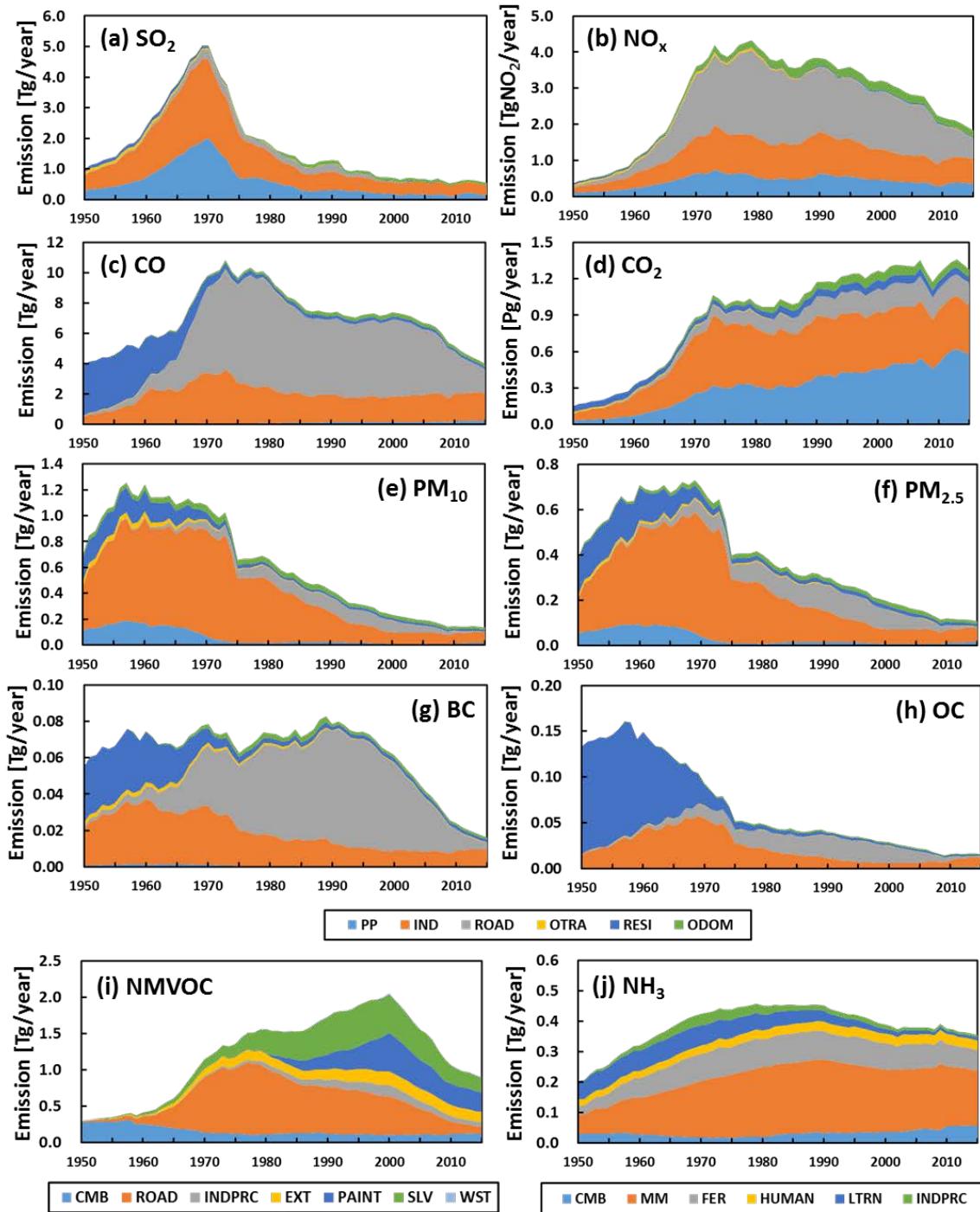


Figure S5: Emissions of (a)  $\text{SO}_2$ , (b)  $\text{NO}_x$ , (c)  $\text{CO}$ , (d)  $\text{CO}_2$ , (e)  $\text{PM}_{10}$ , (f)  $\text{PM}_{2.5}$ , (g)  $\text{BC}$ , (h)  $\text{OC}$ , (i)  $\text{NMVOC}$ , and (j)  $\text{NH}_3$  from major sectors in Japan from 1950 to 2015.

## Japan Fuel

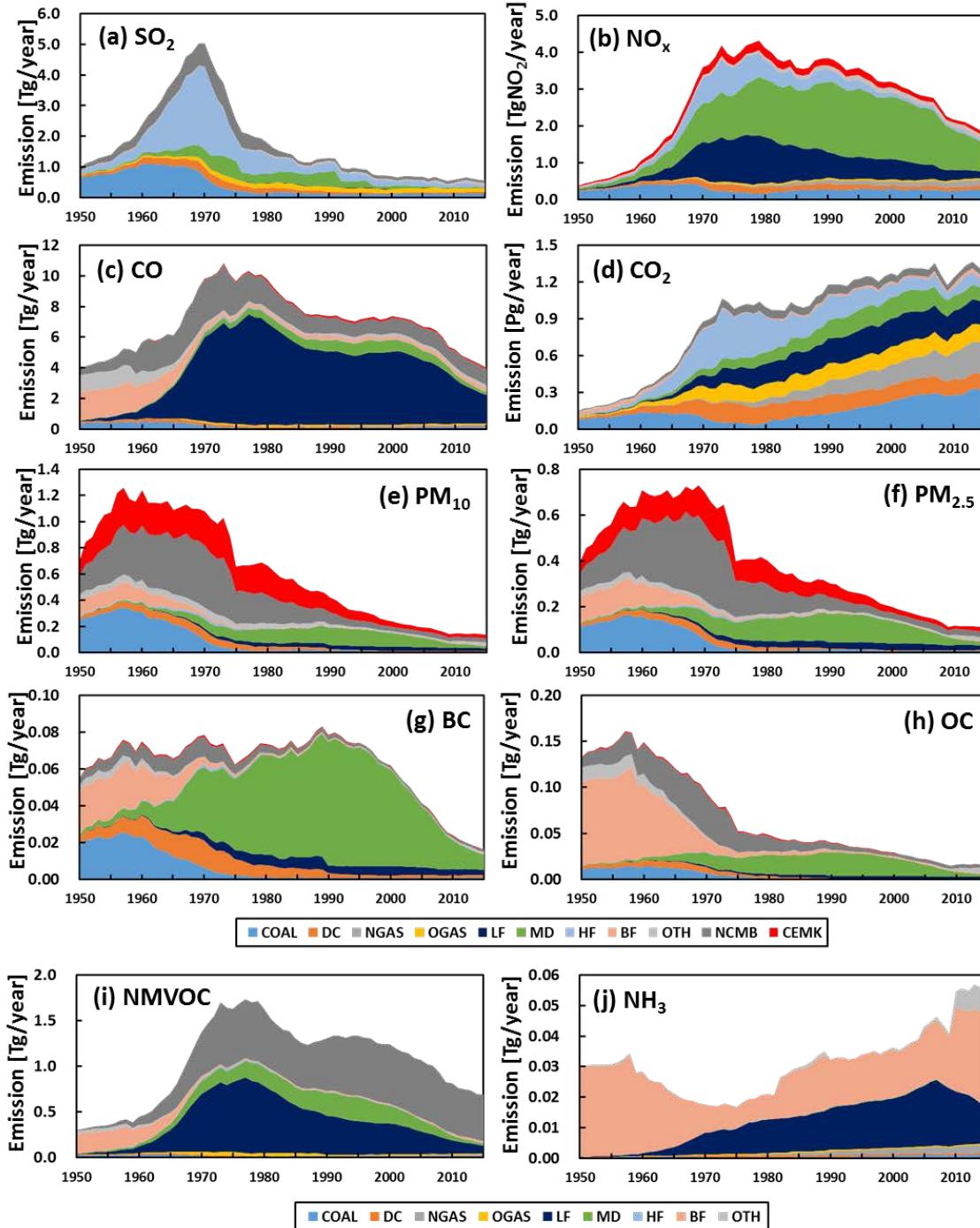


Figure S6: Emissions of (a)  $\text{SO}_2$ , (b)  $\text{NO}_x$ , (c) CO, (d)  $\text{CO}_2$ , (e)  $\text{PM}_{10}$ , (f)  $\text{PM}_{2.5}$ , (g) BC, (h) OC, (i) NMVOC, and (j)  $\text{NH}_3$  from each fuel type in Japan from 1950 to 2015.

## SEA Sector

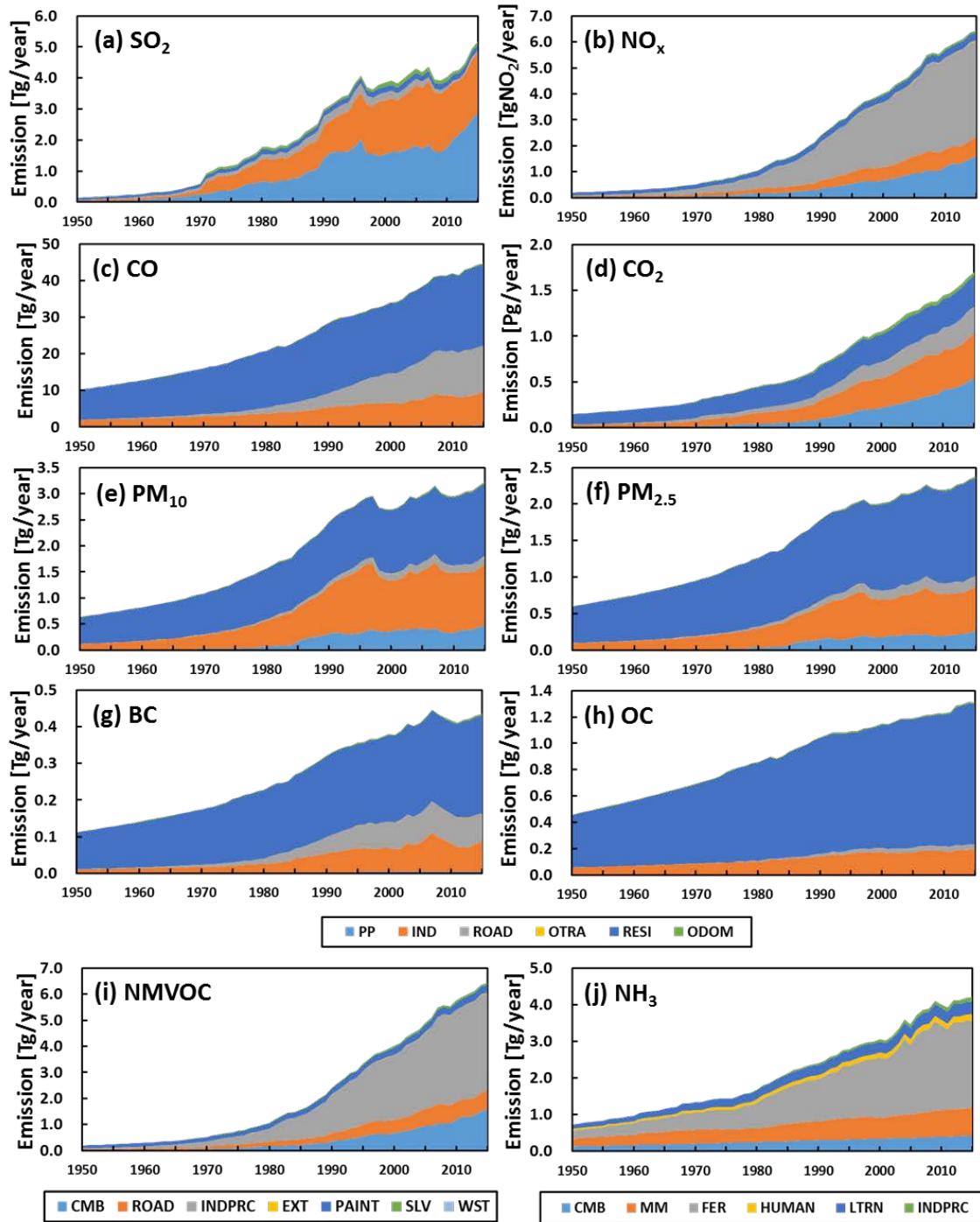


Figure S7: Emissions of (a)  $\text{SO}_2$ , (b)  $\text{NO}_x$ , (c)  $\text{CO}$ , (d)  $\text{CO}_2$ , (e)  $\text{PM}_{10}$ , (f)  $\text{PM}_{2.5}$ , (g)  $\text{BC}$ , (h)  $\text{OC}$ , (i)  $\text{NMVOC}$ , and (j)  $\text{NH}_3$  from major sectors in SEA from 1950 to 2015.

## SEA Fuel

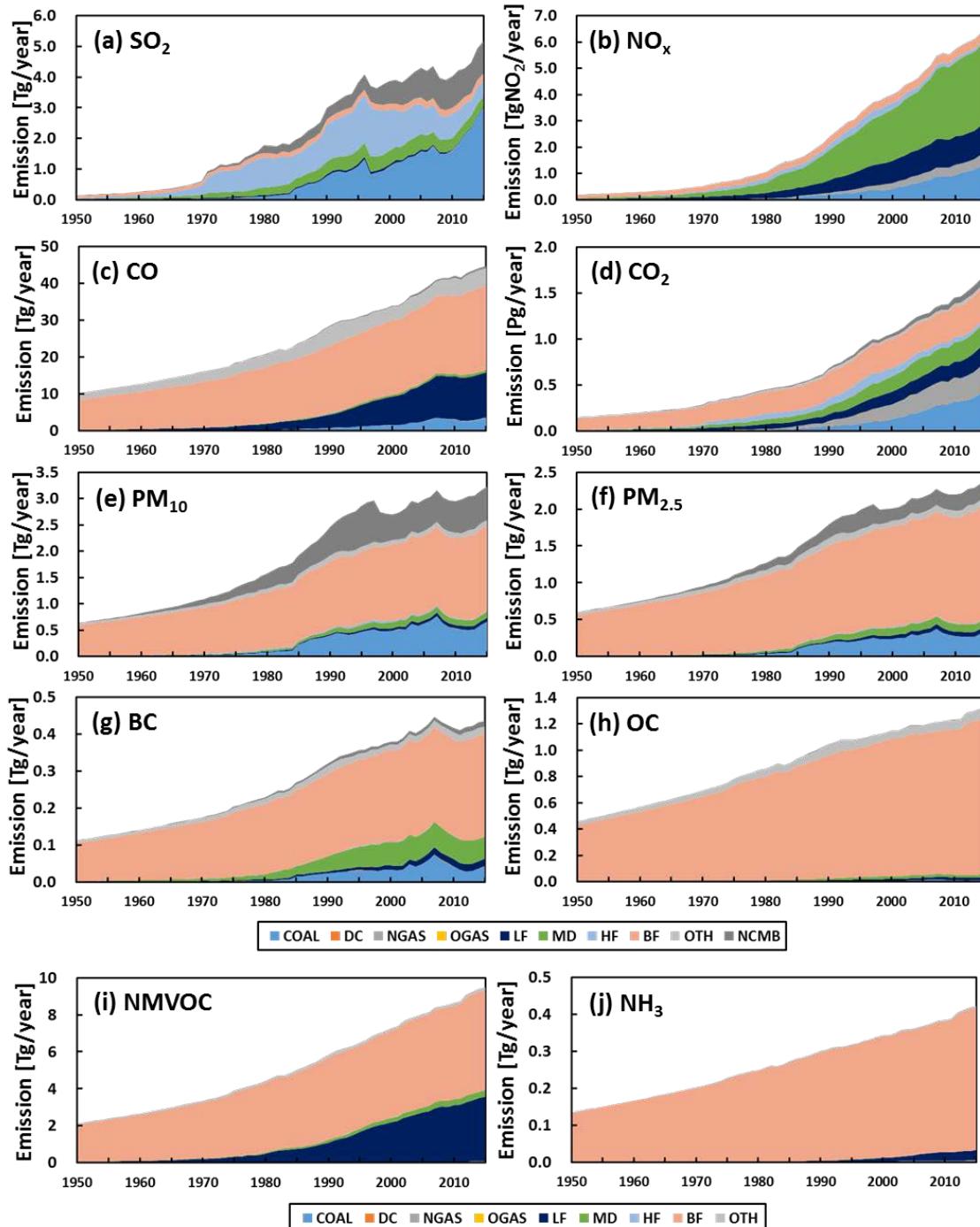
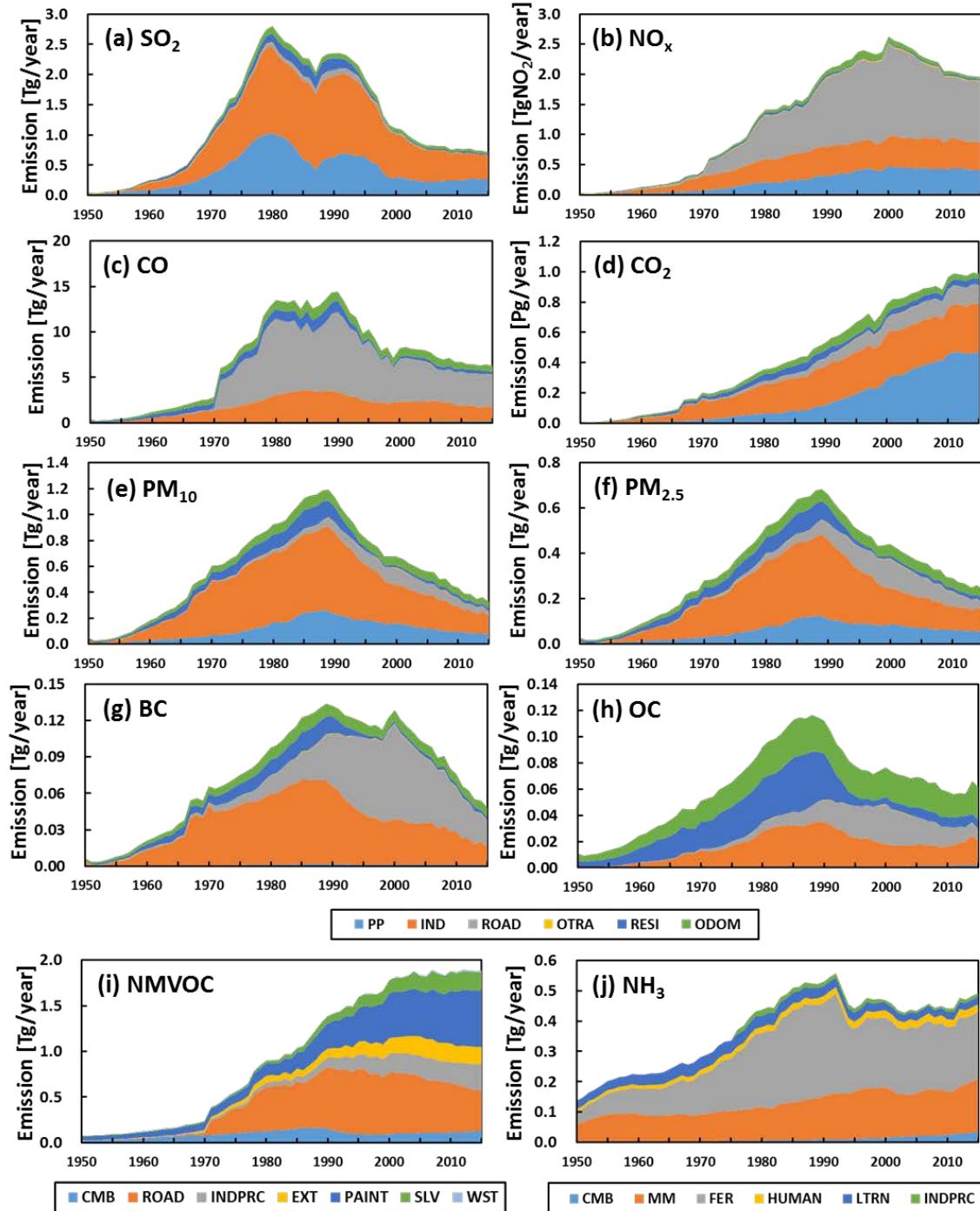


Figure S8: Emissions of (a)  $\text{SO}_2$ , (b)  $\text{NO}_x$ , (c) CO, (d)  $\text{CO}_2$ , (e)  $\text{PM}_{10}$ , (f)  $\text{PM}_{2.5}$ , (g) BC, (h) OC, (i) NMVOC, and (j)  $\text{NH}_3$  from each fuel type in SEA from 1950 to 2015.

## OEA Sector



60 Figure S9: Emissions of (a)  $\text{SO}_2$ , (b)  $\text{NO}_x$ , (c) CO, (d)  $\text{CO}_2$ , (e)  $\text{PM}_{10}$ , (f)  $\text{PM}_{2.5}$ , (g) BC, (h) OC, (i) NMVOC, and (j)  $\text{NH}_3$  from major sectors in OEA from 1950 to 2015.

## OEA Fuel

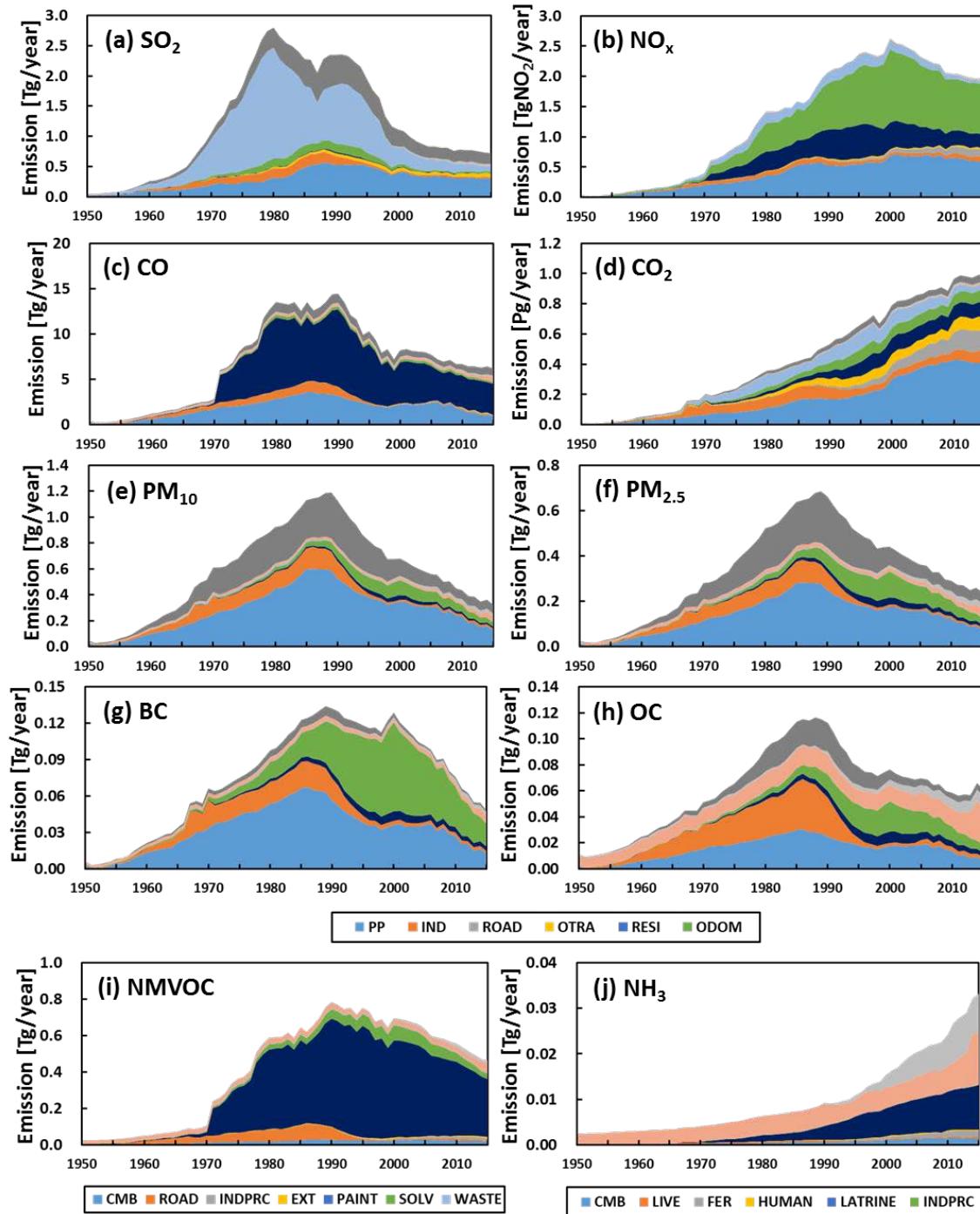


Figure S10: Emissions of (a)  $\text{SO}_2$ , (b)  $\text{NO}_x$ , (c) CO, (d)  $\text{CO}_2$ , (e)  $\text{PM}_{10}$ , (f)  $\text{PM}_{2.5}$ , (g) BC, (h) OC, (i) NMVOC, and (j)  $\text{NH}_3$  from each fuel type in OEA from 1950 to 2015.

## OSA Sector

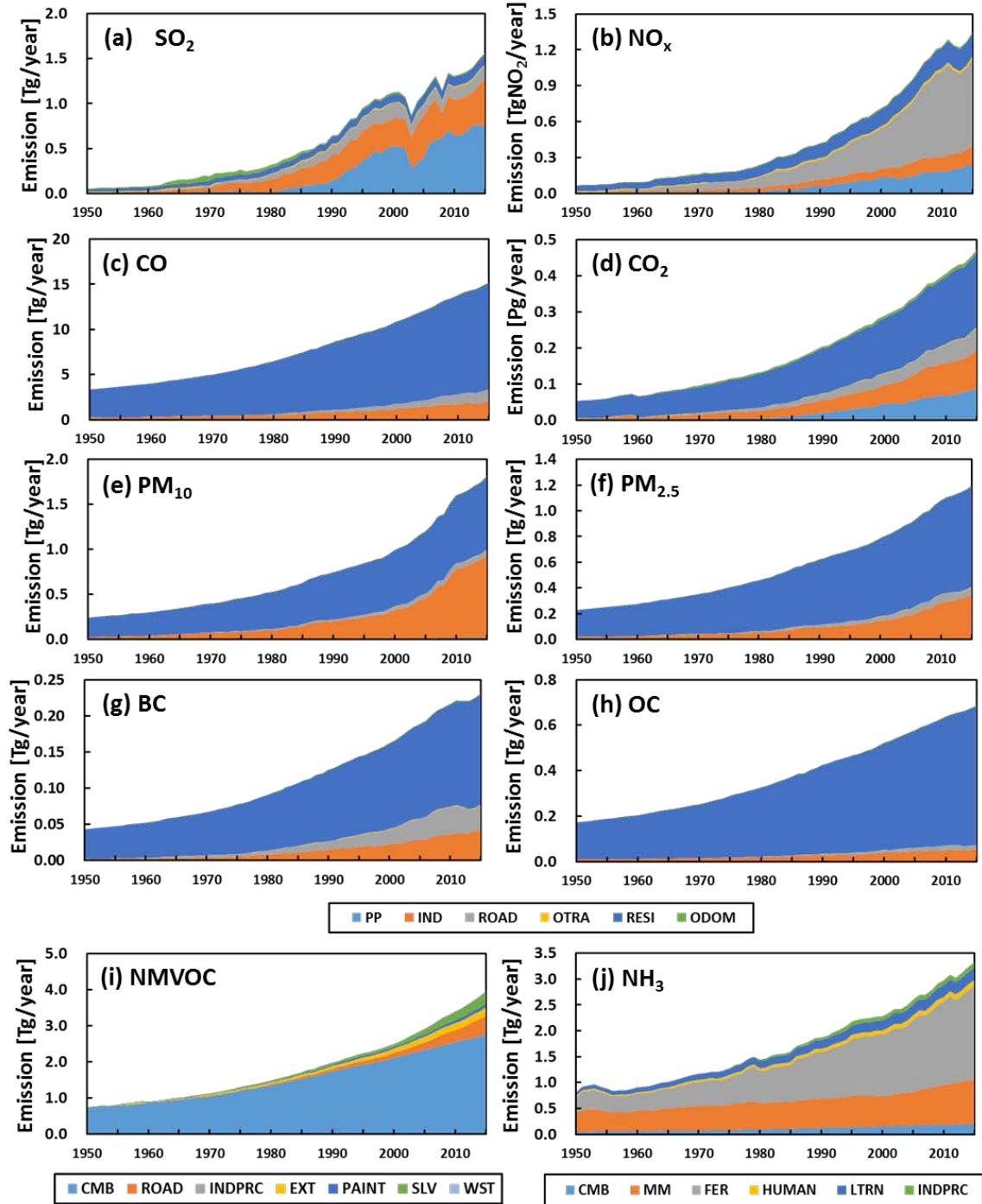


Figure S11: Emissions of (a)  $\text{SO}_2$ , (b)  $\text{NO}_x$ , (c)  $\text{CO}$ , (d)  $\text{CO}_2$ , (e)  $\text{PM}_{10}$ , (f)  $\text{PM}_{2.5}$ , (g)  $\text{BC}$ , (h)  $\text{OC}$ , (i)  $\text{NMVOC}$ , and (j)  $\text{NH}_3$  from major sectors in OSA from 1950 to 2015.

## OSA Fuel

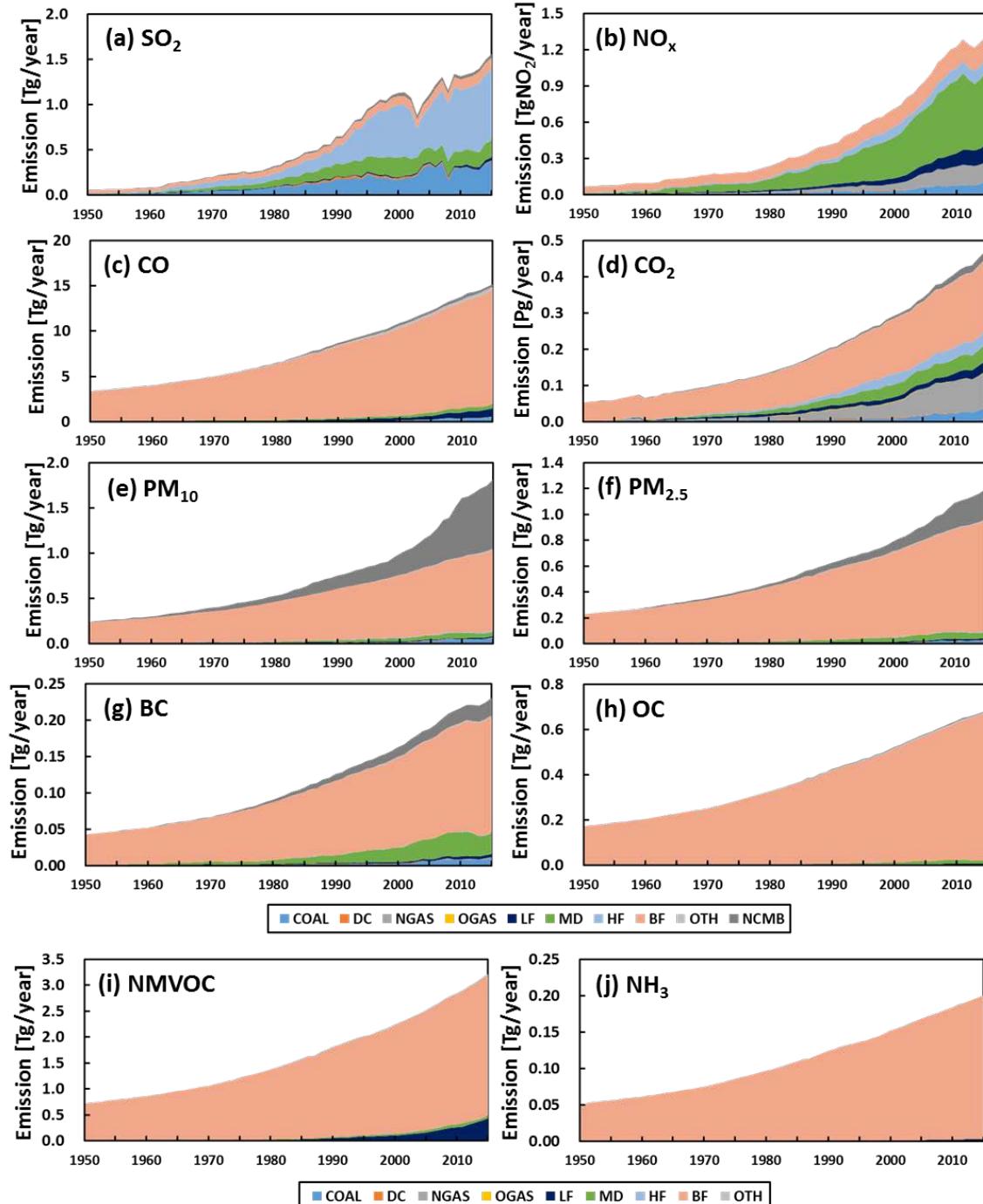
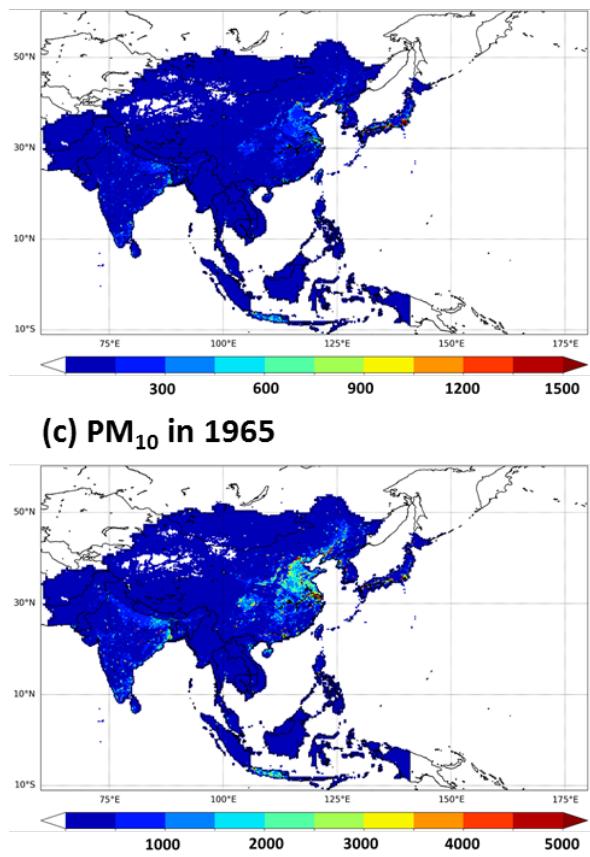
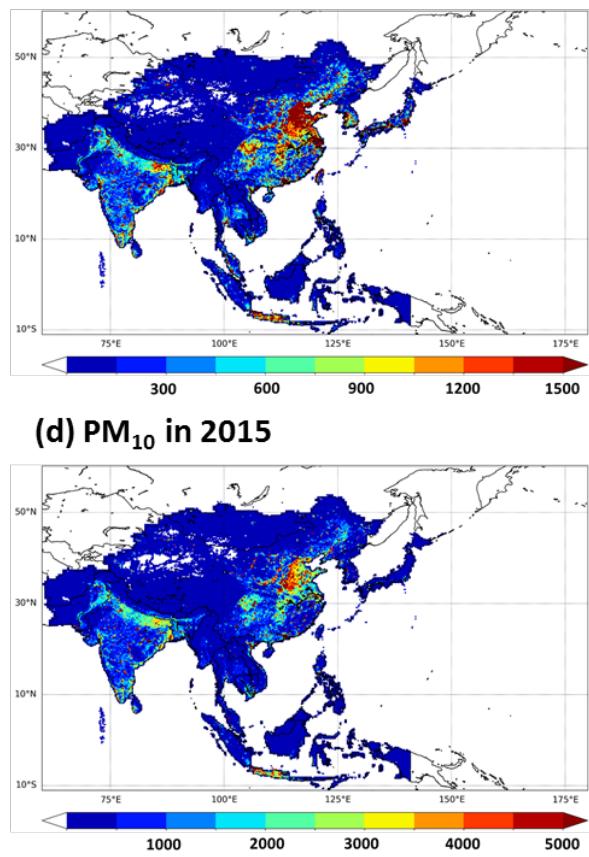


Figure S12: Emissions of (a) SO<sub>2</sub>, (b) NO<sub>x</sub>, (c) CO, (d) CO<sub>2</sub>, (e) PM<sub>10</sub>, (f) PM<sub>2.5</sub>, (g) BC, (h) OC, (i) NMVOC, and (j) NH<sub>3</sub> from each fuel type in OSA from 1950 to 2015.

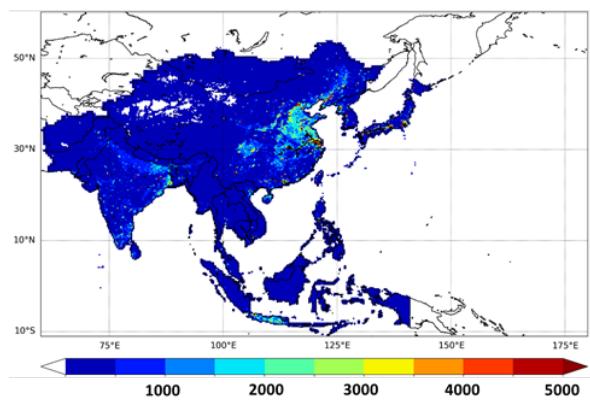
**(a) CO<sub>2</sub> in 1965**



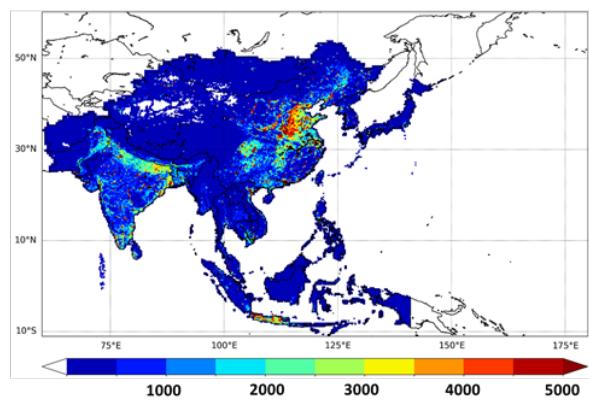
**(b) CO<sub>2</sub> in 2015**



**(c) PM<sub>10</sub> in 1965**



**(d) PM<sub>10</sub> in 2015**



**Figure S13:** Grid maps of annual emissions of (a, c) CO<sub>2</sub> (kt year<sup>-1</sup> per grid cell) and (b, d) PM<sub>10</sub> (t year<sup>-1</sup> per grid cell) in 1965 (left) and 2015 (right).

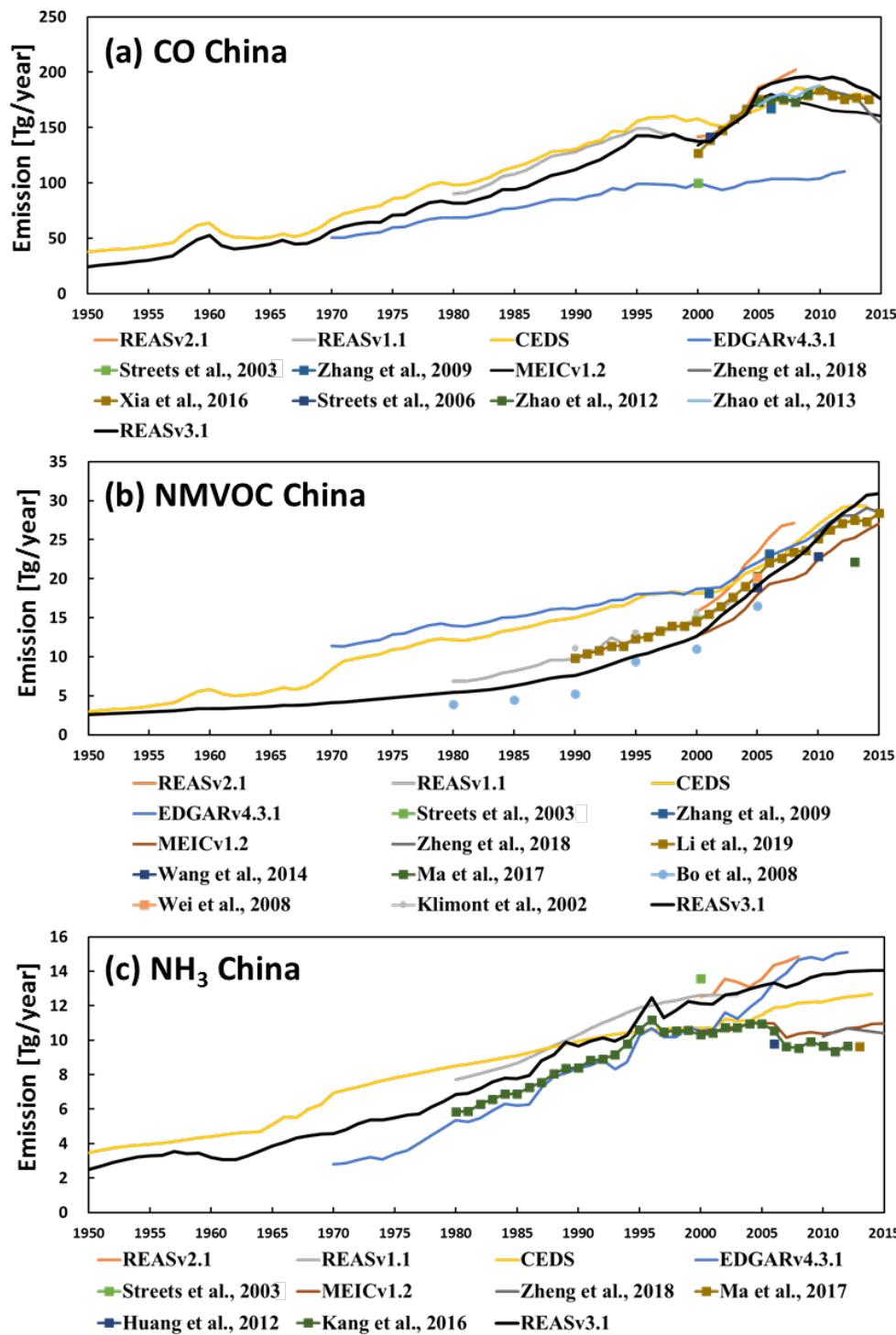


Figure S14: Comparison of (a) CO, (b) NMVOC, (c) NH<sub>3</sub>, (d) PM<sub>10</sub>, (e) PM<sub>2.5</sub> and (f) BC emissions in China between REASv3.1 and other studies.

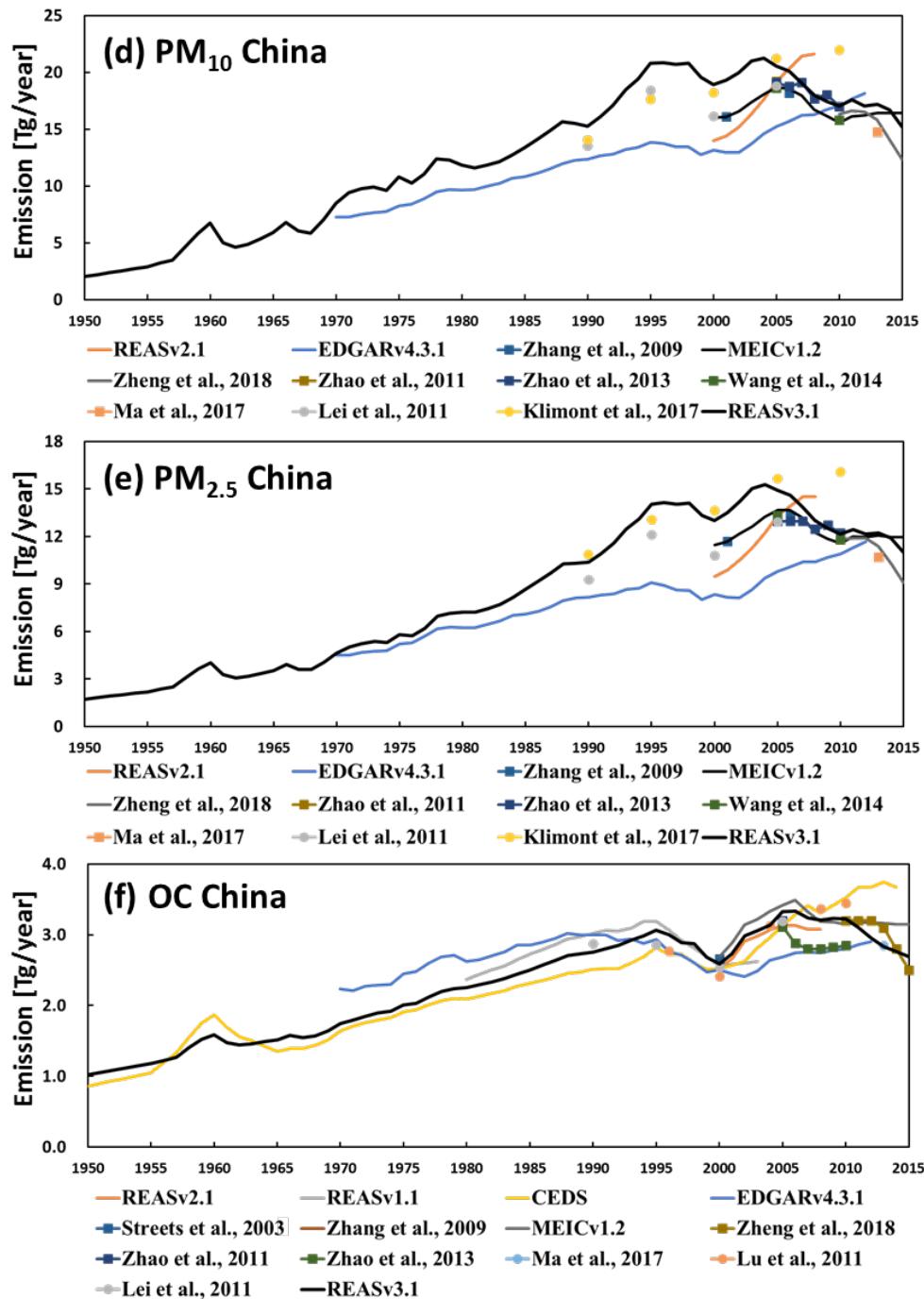


Figure S14: Continued.

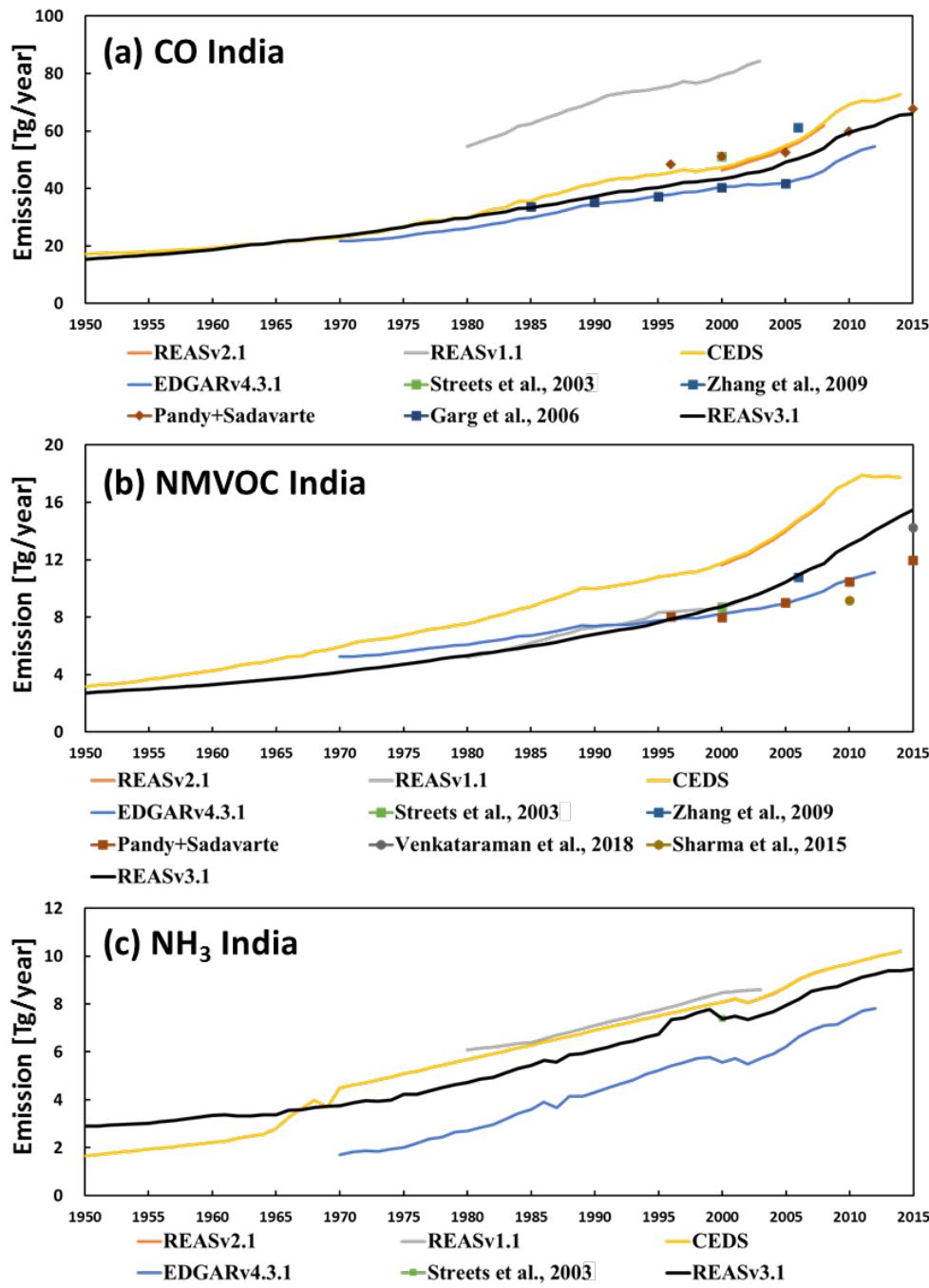


Figure S15: Comparison of (a) CO, (b) NMVOC, (c) NH<sub>3</sub>, (d) PM<sub>10</sub>, (e) PM<sub>2.5</sub> and (f) OC emissions in India between REASv3.1 and other studies.

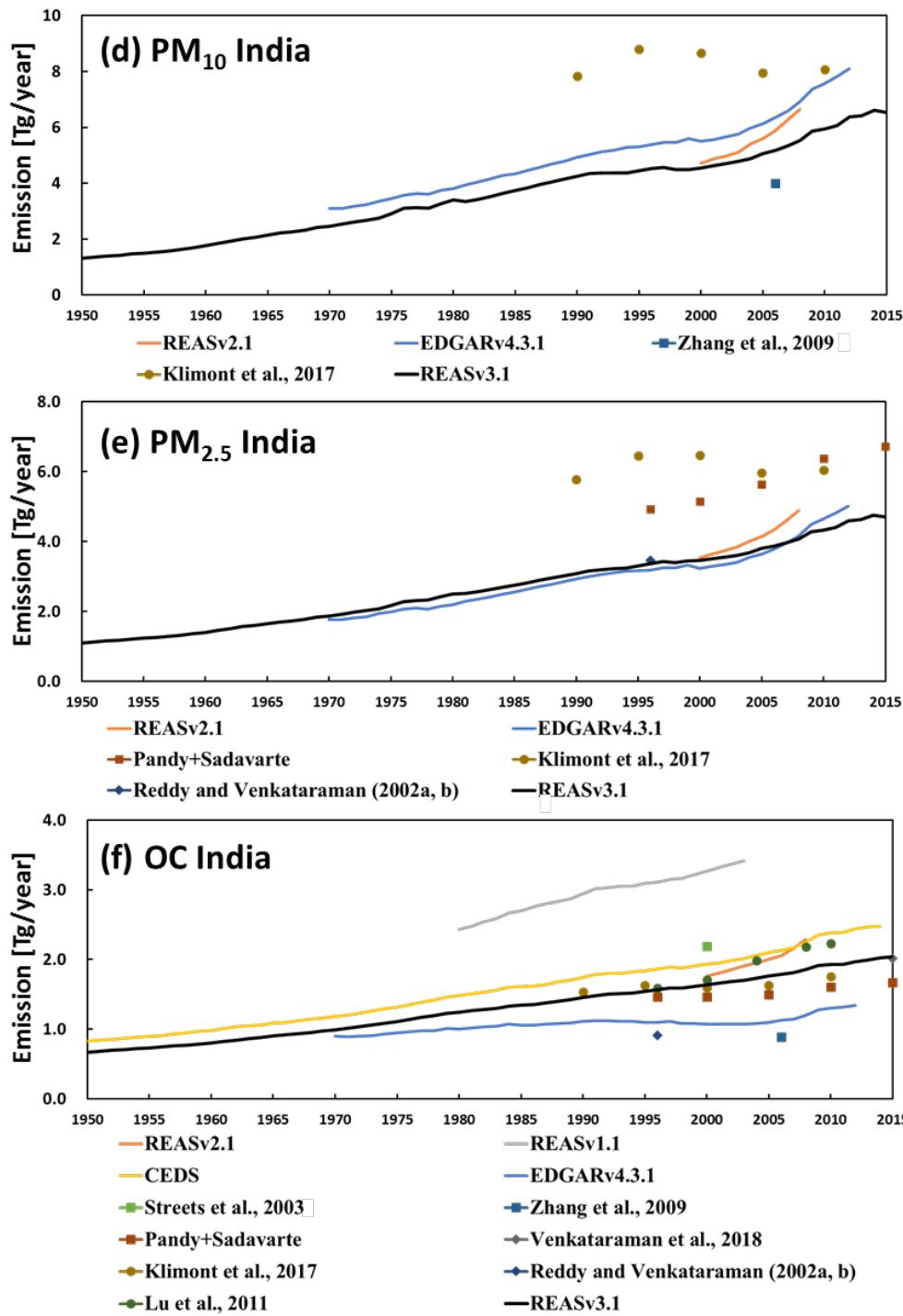


Figure S15: Continued.

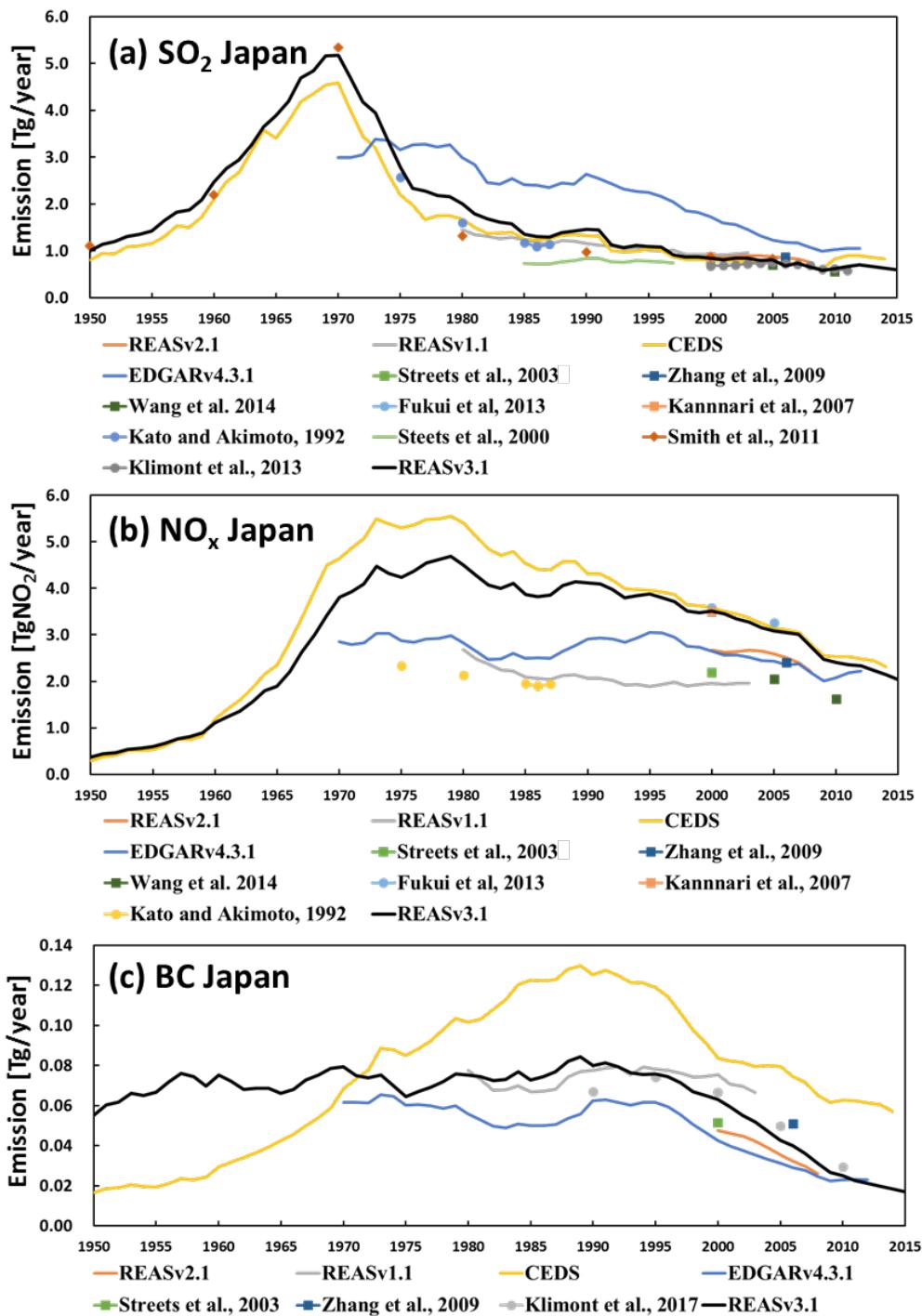


Figure S16: Comparison of (a) SO<sub>2</sub>, (b) NO<sub>x</sub>, (c) BC, (d) CO, (e) NMVOC, (f) NH<sub>3</sub>, (g) PM<sub>10</sub>, (h) PM<sub>2.5</sub> and (i) OC emissions in Japan between REASv3.1 and other studies.

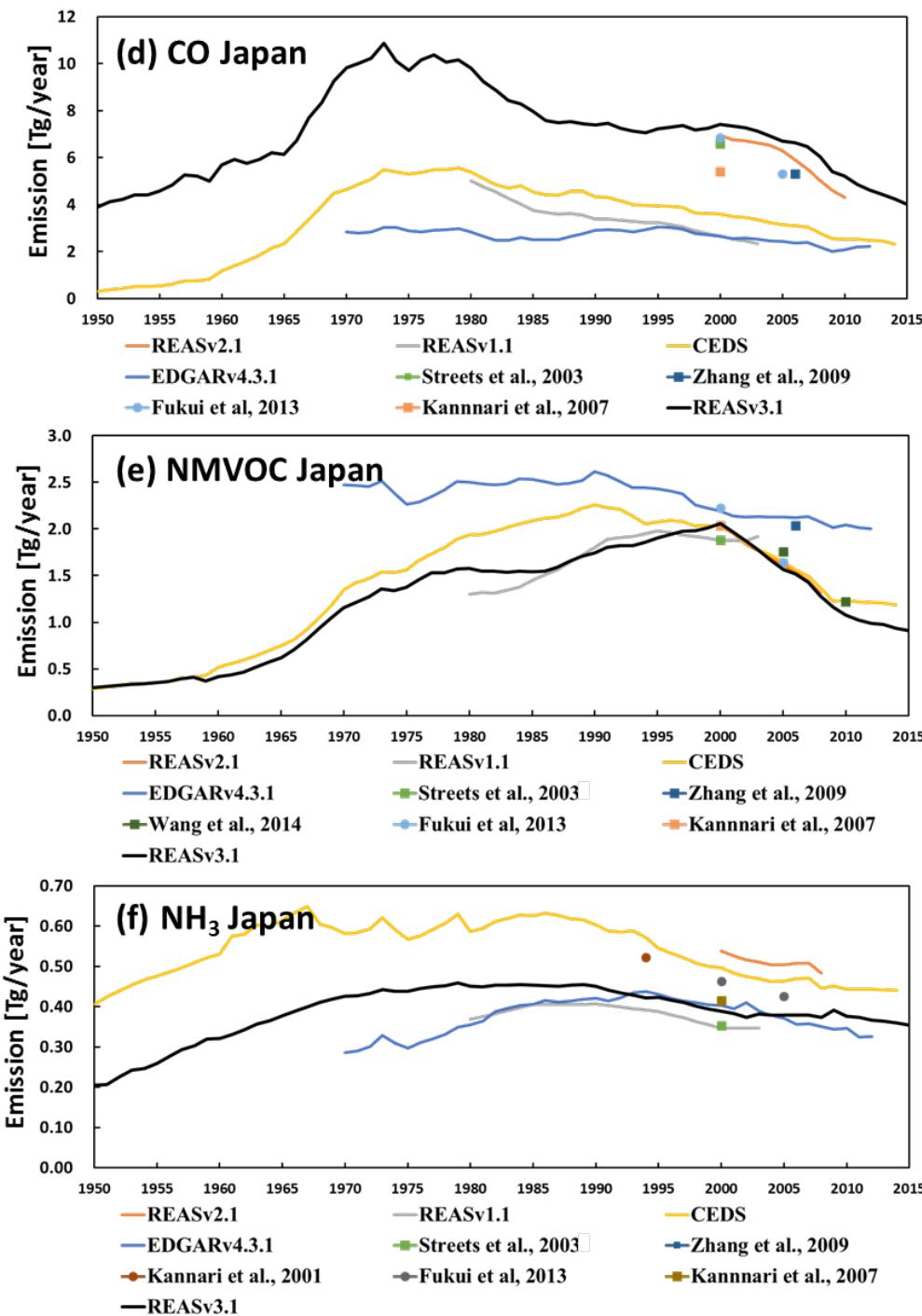
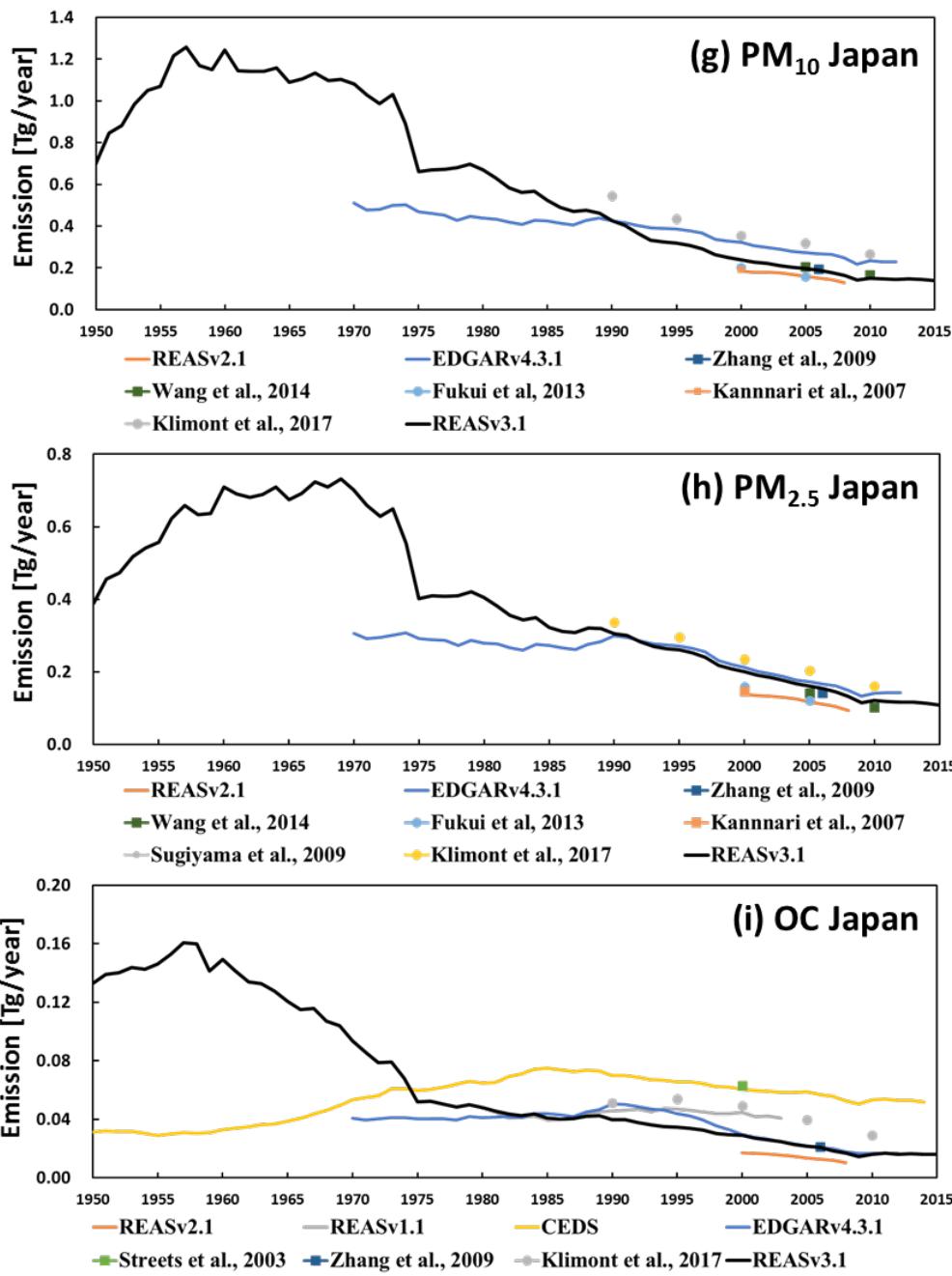


Figure S16: Continued.



95 Figure S16: Continued.

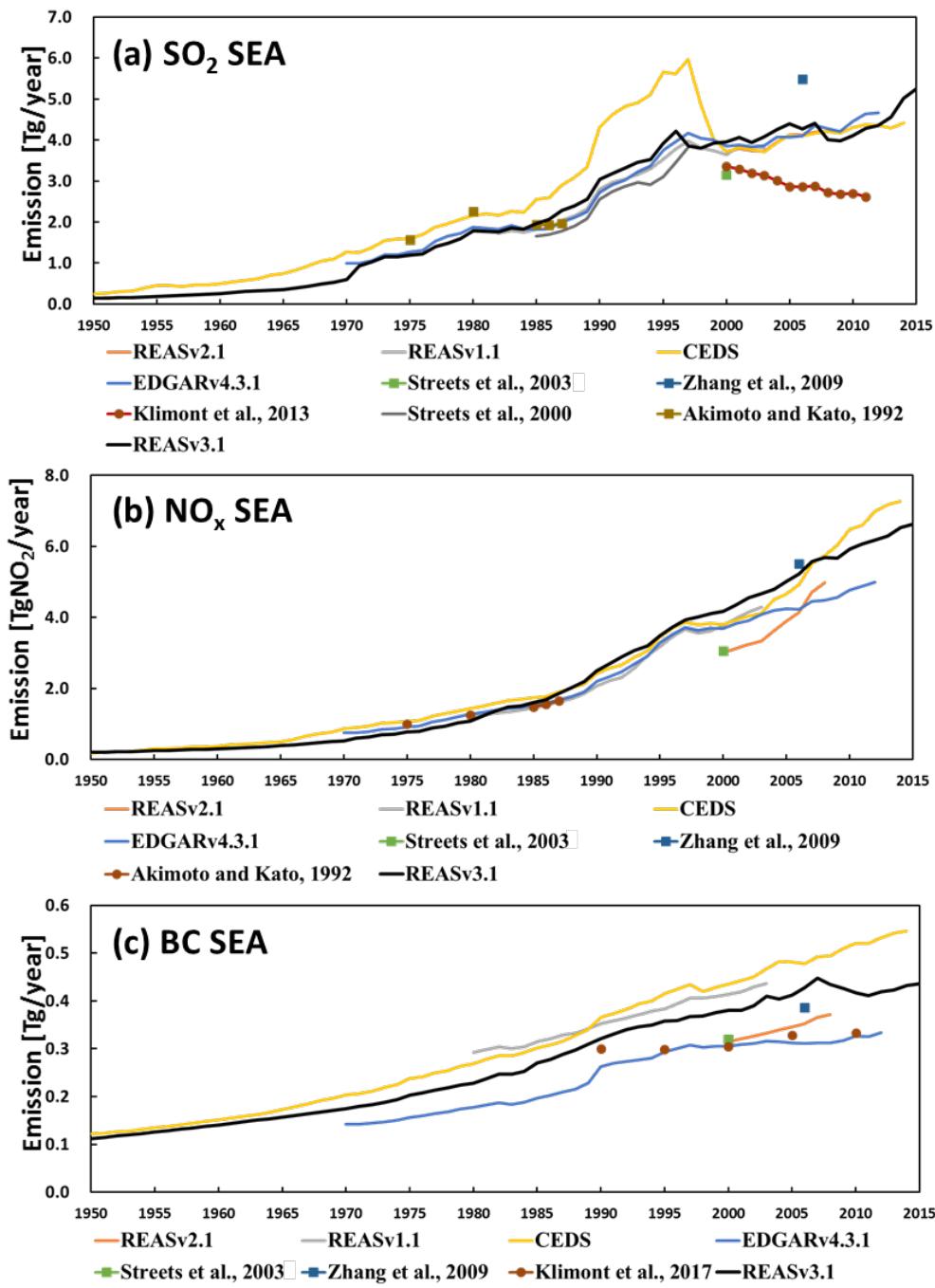


Figure S17: Comparison of (a) SO<sub>2</sub>, (b) NO<sub>x</sub>, (c) BC, (d) CO, (e) NMVOC, (f) NH<sub>3</sub>, (g) PM<sub>10</sub>, (h) PM<sub>2.5</sub> and (i) OC emissions in SEA between REASv3.1 and other studies.

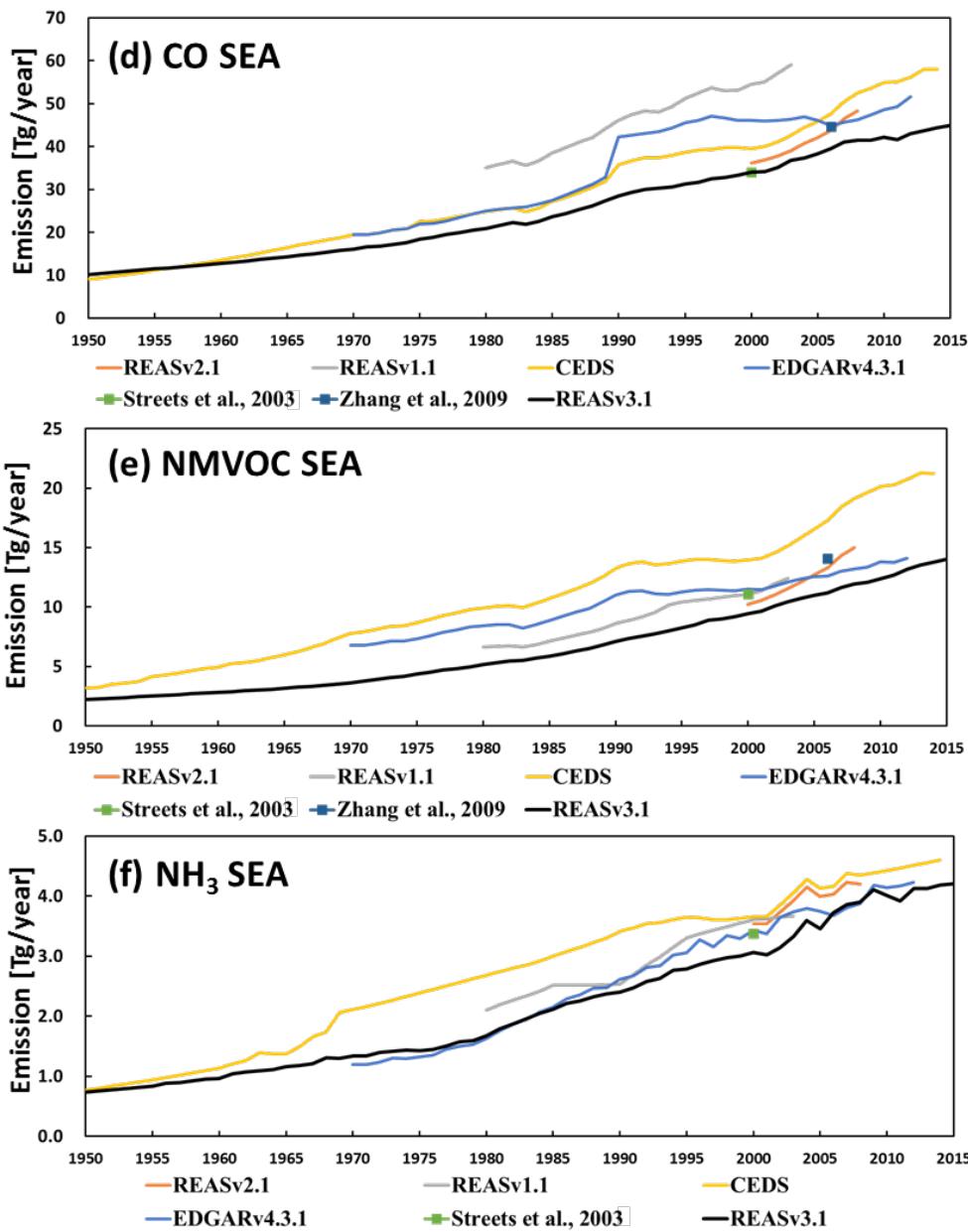
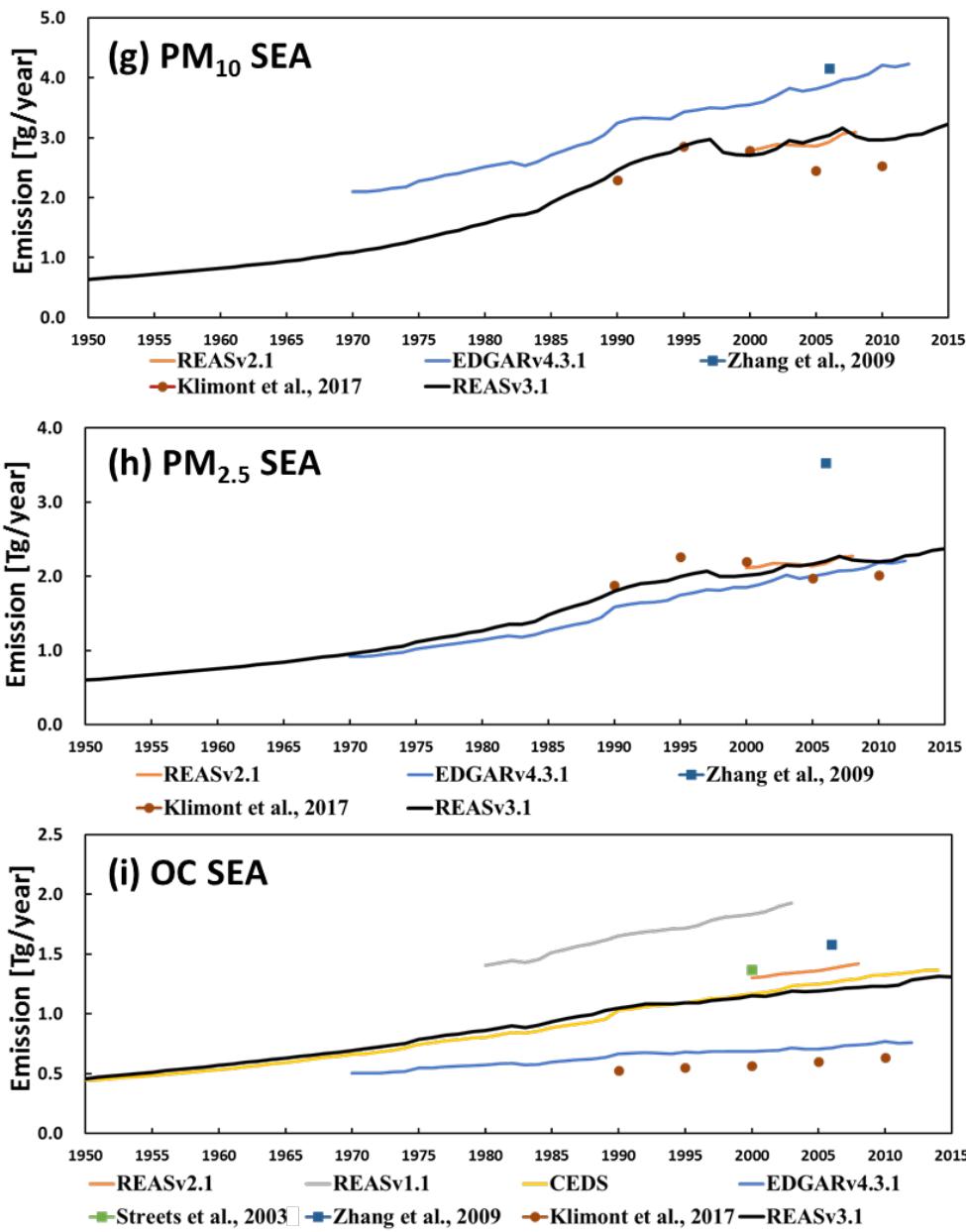
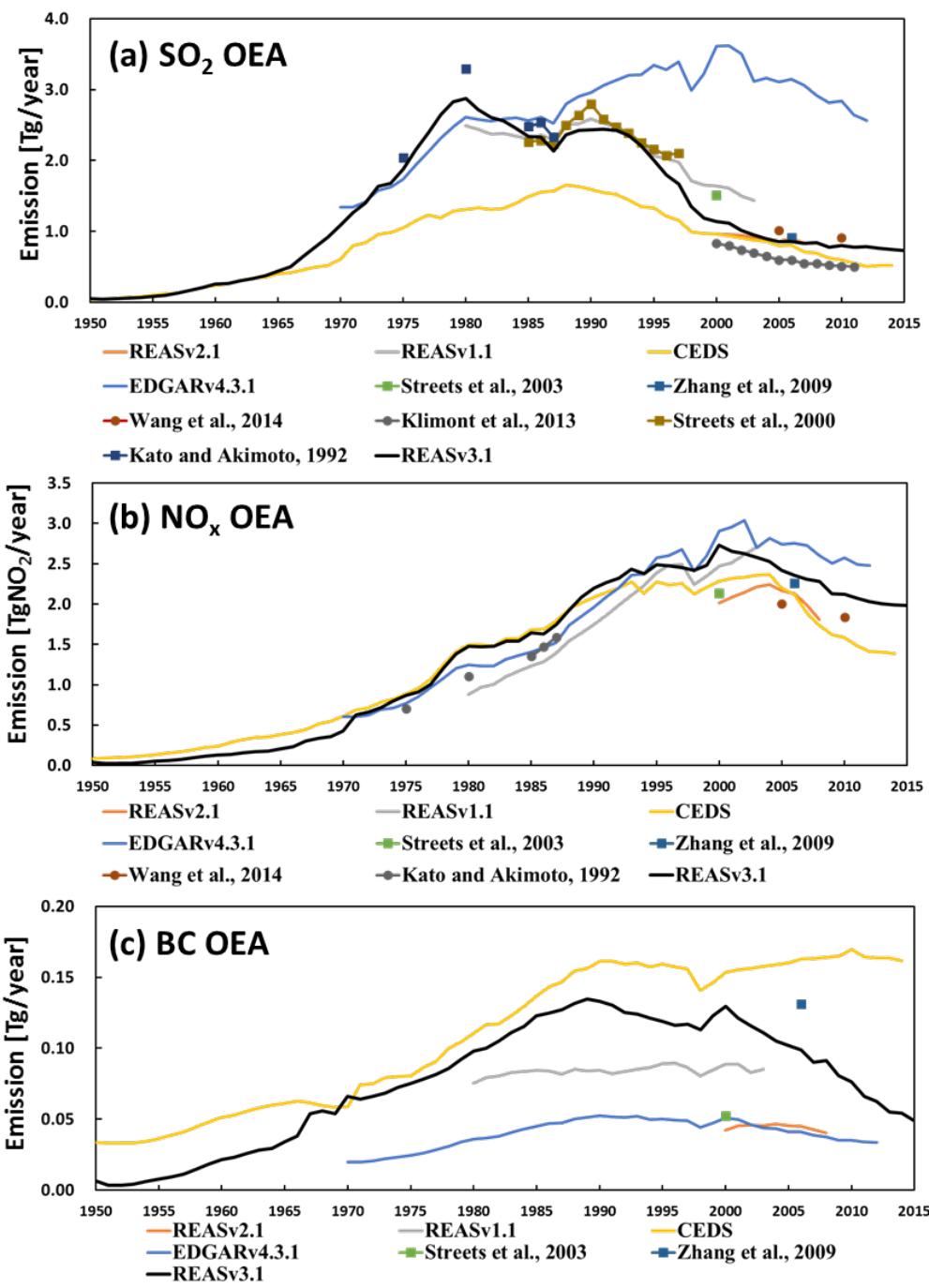


Figure S17: Continued.



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Figure S17: Continued.



110 Figure S18: Comparison of (a) SO<sub>2</sub>, (b) NO<sub>x</sub>, (c) BC, (d) CO, (e) NMVOC, (f) NH<sub>3</sub>, (g) PM<sub>10</sub>, (h) PM<sub>2.5</sub> and (i) OC emissions in OEA between REASv3.1 and other studies.

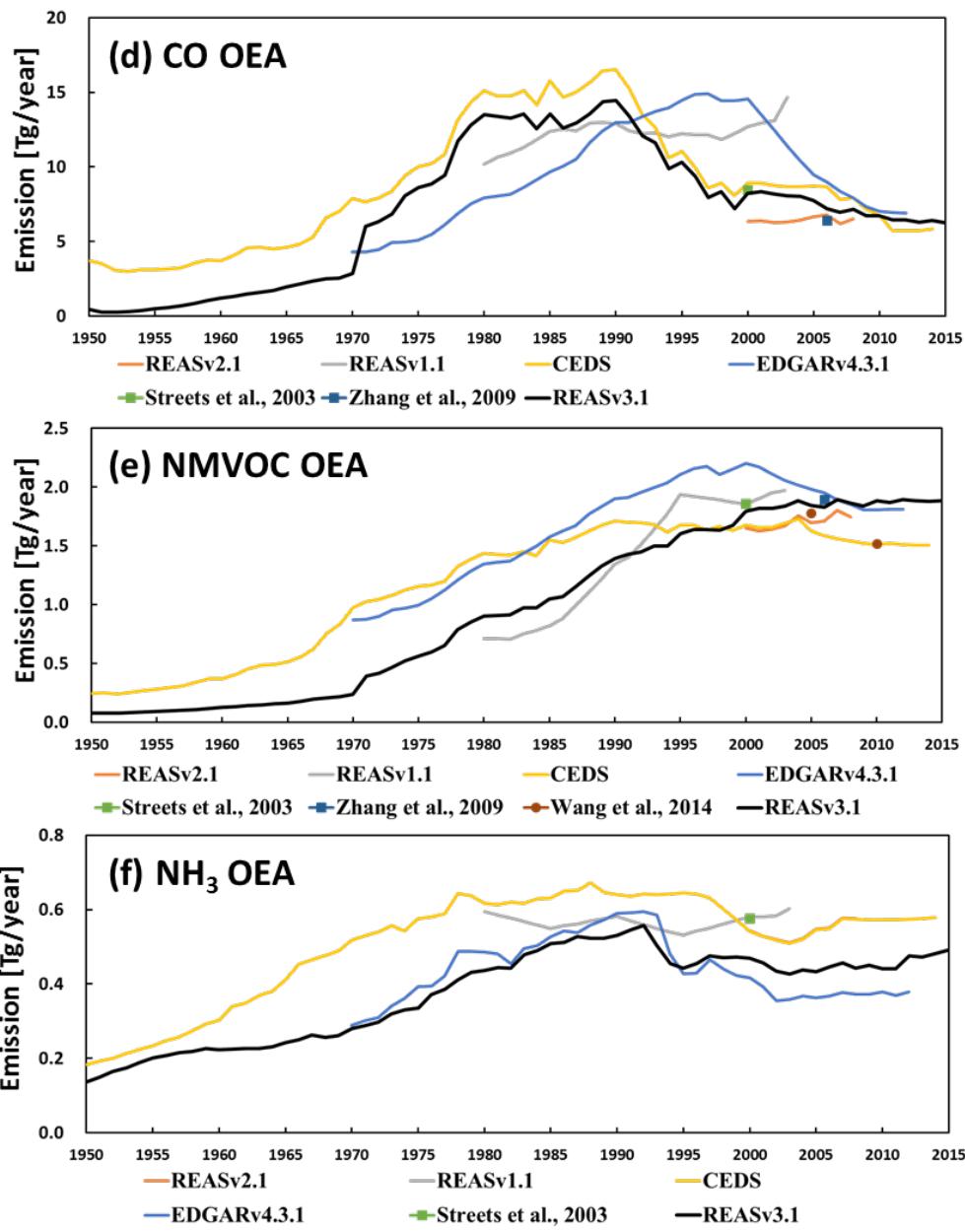
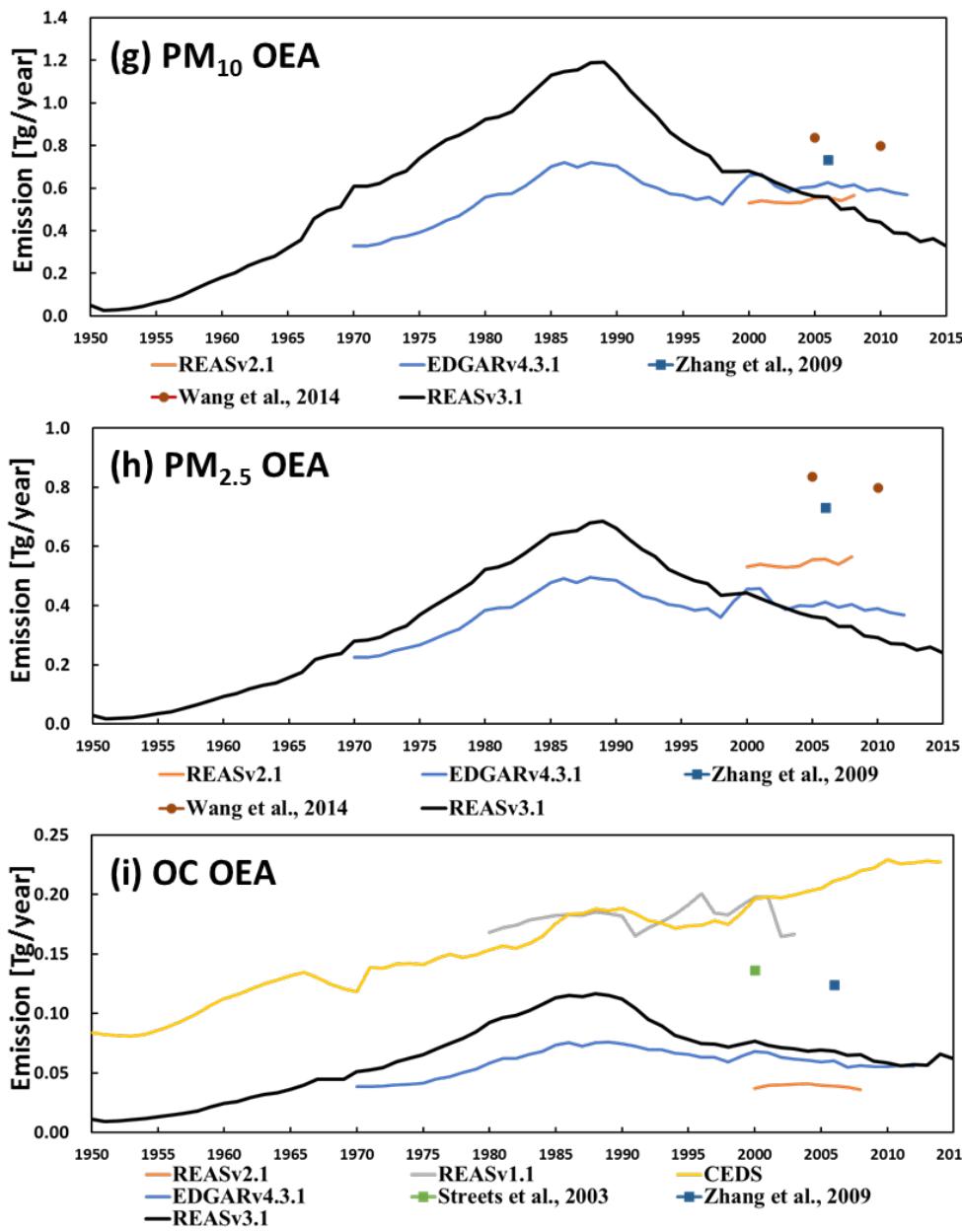


Figure S18: Continued.



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Figure S18: Continued.

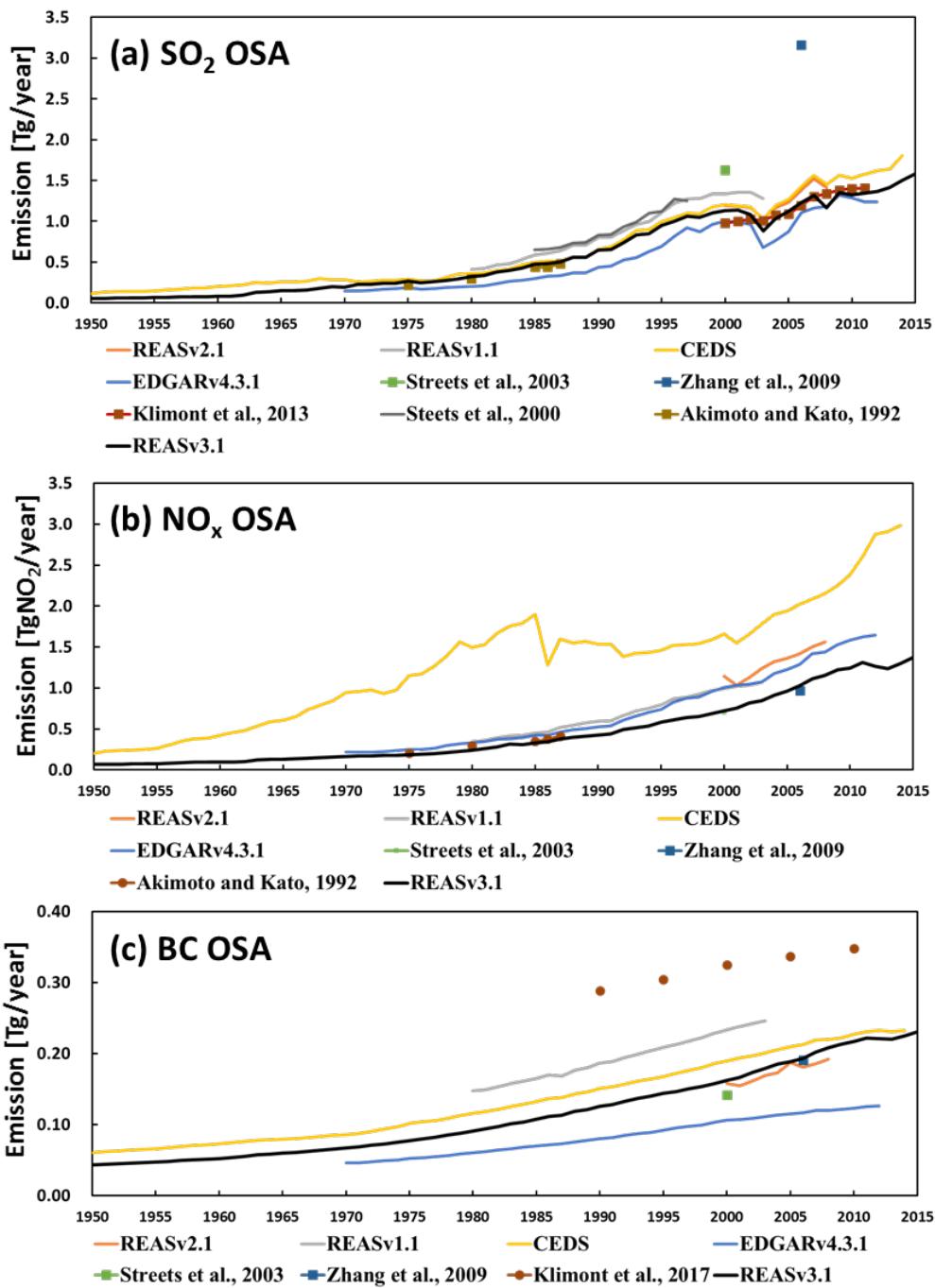


Figure S19: Comparison of (a)  $\text{SO}_2$ , (b)  $\text{NO}_x$ , (c) BC, (d) CO, (e) NMVOC, (f)  $\text{NH}_3$ , (g)  $\text{PM}_{10}$ , (h)  $\text{PM}_{2.5}$  and (i) OC emissions in OSA between REASv3.1 and other studies.

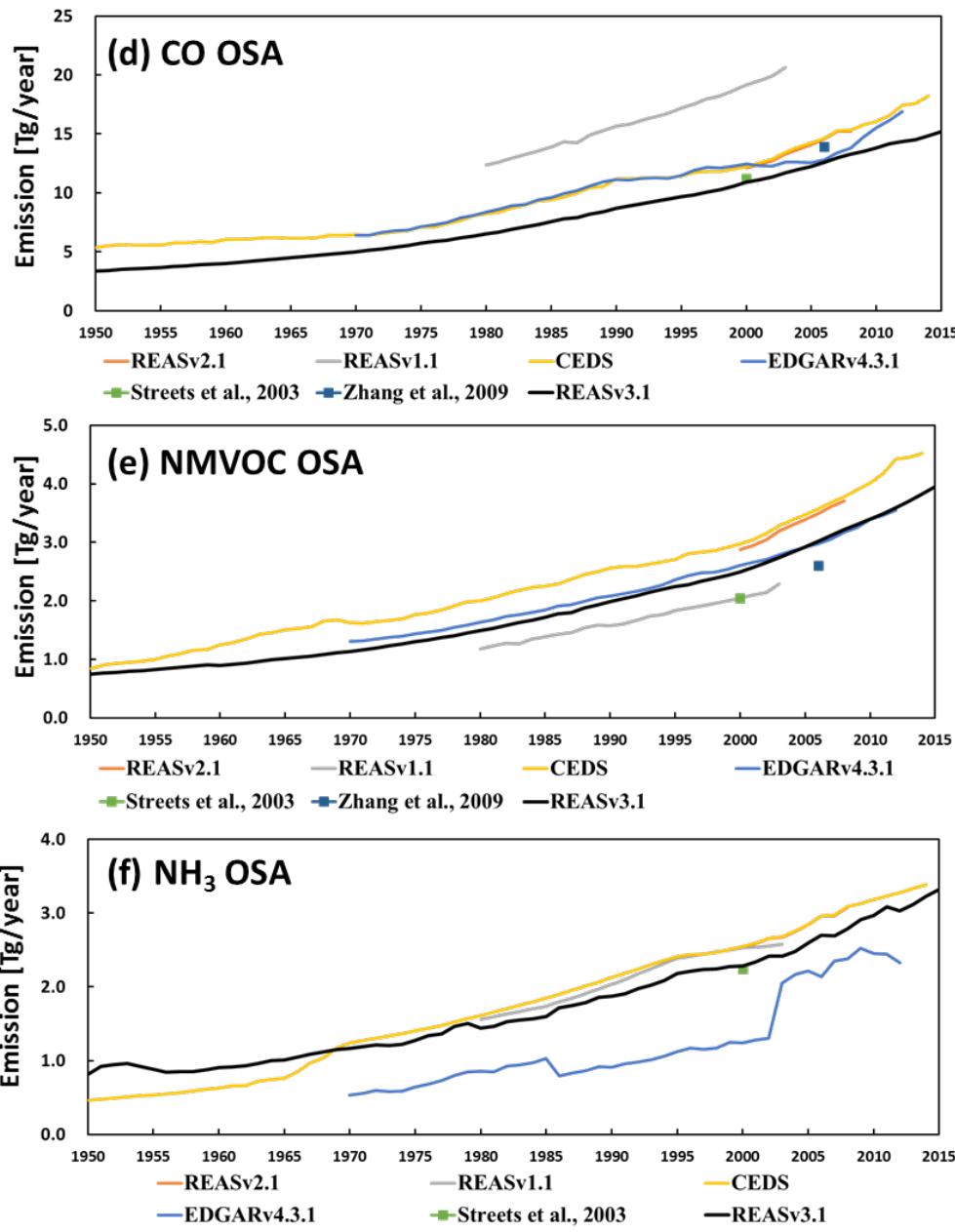
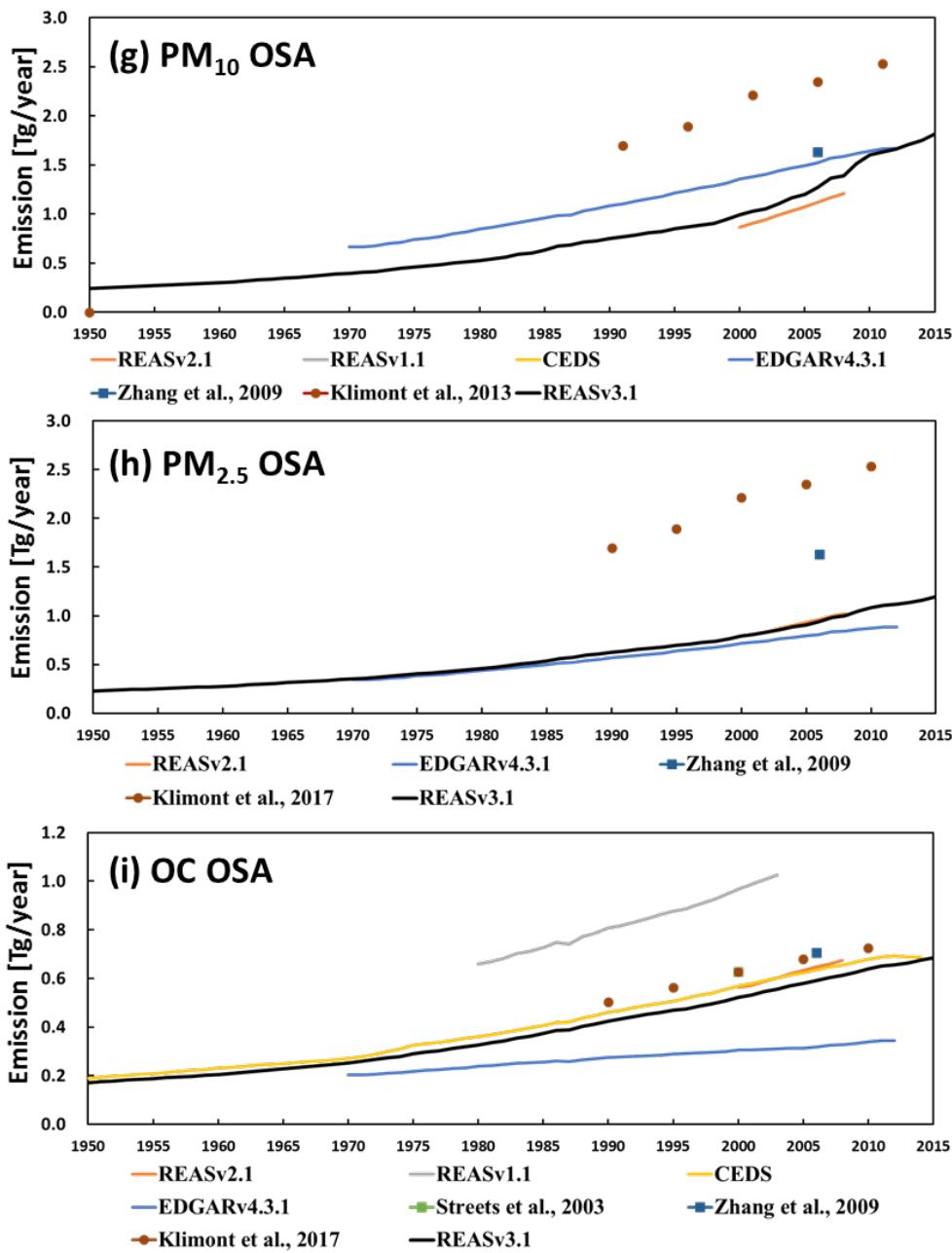
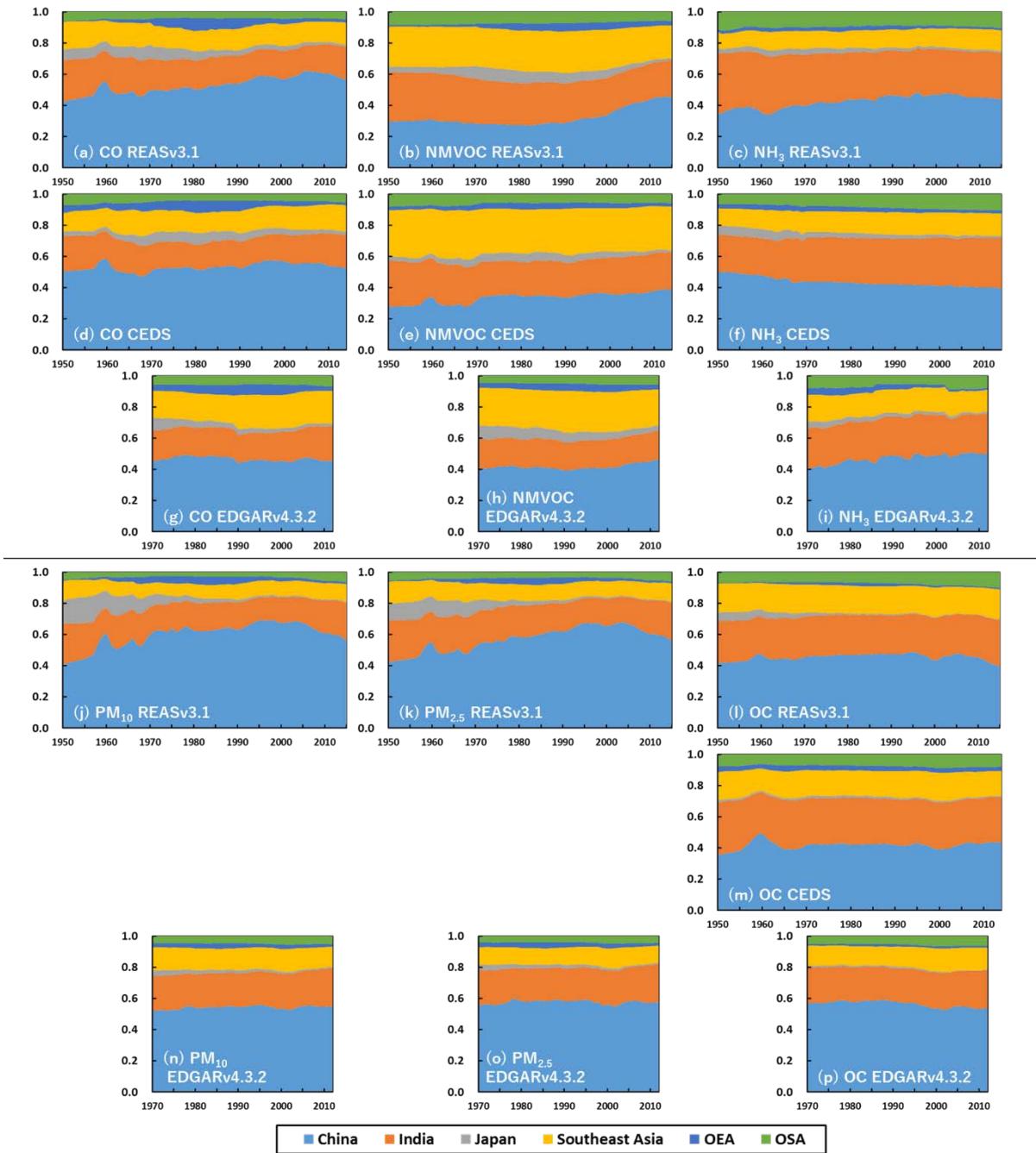


Figure S19: Continued.



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Figure S19: Continued.



130 **Figure S20: Comparison of trends of relative ratios of emissions from China, India, Japan, Southeast Asia, OEA, and OSA for (a, d, g) CO, (b, e, h) NMVOC, (c, f, i) NH<sub>3</sub>, (j, n) PM<sub>10</sub>, (k, o) PM<sub>2.5</sub>, and (l, m, p) OC among (a, b, c, j, k, l) REASv3.1, (d, e, f, m) CEDS, and (g, h, i, n, o, p) EDGARv4.3.2.**

## Supplement Tables

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**Table S1** provides target countries and sub-regions in REASv3.1 with their codes used in the manuscript and country and regional emission table data for major sectors and those for major fuel types available from a data download site of REAS (<http://www.nies.go.jp/REAS/>).

**Table S2** shows uncertainties of emissions of SO<sub>2</sub>, NO<sub>x</sub>, CO, CO<sub>2</sub>, PM<sub>10</sub>, PM<sub>2.5</sub>, BC, and OC for each sector in China, India,  
140 Japan, Southeast Asia (SEA), East Asia other than China and Japan (OEA), and South Asia other than India (OSA) in  
2015, 1985, and 1955.

**Table S3** presents uncertainties of emissions of NMVOC for each sector in China, India, Japan, SEA, OEA, and OSA in  
2015, 1985, and 1955.

**Table S4** provides uncertainties of emissions of NH<sub>3</sub> for each sector in China, India, Japan, SEA, OEA, and OSA in  
145 2015, 1985, and 1955.

**Table S1: Countries and sub-regions included in REAS 3.1.**

Region	Country/Sub-region	Code
China (CHN)	China/Whole Country	CHNWC
	China/Beijing	CHNBJ
	China/Tianjin	CHNTJ
	China/Hebei	CHNHE
	China/Shanxi	CHNSX
	China/Inner Mongolia	CHNNM
	China/Liaoning	CHNLN
	China/Jilin	CHN JL
	China/Heilongjiang	CHNHL
	China/Shanghai	CHNSH
	China/Jiangsu	CHNJS
	China/Zhejiang	CHNZJ
	China/Anhui	CHNAH
	China/Fujian	CHNFJ
	China/Jiangxi	CHNJX
	China/Shandong	CHNSD
	China/Henan	CHNHA
	China/Hubei	CHNHB
	China/Hunan	CHNHN
	China/Guangdong	CHNGD
	China/Guangxi	CHNGX
	China/Hainan	CHNHI
	China/Chongqing	CHNCQ
	China/Sichuan	CHNSC
	China/Guizhou	CHNGZ
	China/Yunnan	CHNYN
	China/Tibet	CHNXZ
	China/Shaanxi	CHNSN
	China/Gansu	CHNGS
	China/Qinghai	CHNQH
	China/Ningxia	CHNNX
	China/Xinjiang	CHNXJ
	China/Hong Kong	CHNHK
	China/Macau	CHNMC
India (IND)	India/Whole Country	INDWC
	India/Andhra Pradesh	INDAP
	India/Bihar, Jharkhand	INDBJ
	India/North East (Arunachal Pradesh, Assam, Manipur, Meghalaya, Mizoram, Nagaland, Sikkim, Tripura)	INDAN
	India/Gujarat	INDGU
	India/Haryana	INDHA
	India/Karnataka, Goa	INDKG
	India/Kerala	INDKE
	India/Madhya Pradesh, Chhattisgarh	INDMC
	India/Maharashtra	INDMA

	India/Orissa	INDOR
	India/Punjab, Chandigarh	INDPU
	India/Rajasthan	INDRA
	India/Tamil Nadu	INDTN
	India/Utter Pradesh, Uttaranchal	INDUU
	India/West Bengal	INDWB
	India/Himachal Pradesh, Jammu and Kashmir	INDHJ
	India/Delhi	INDDE
Japan (JPN)	Japan/Whole Country	JPNWC
	Japan/Hokkaido-Tohoku	JPNHT
	Japan/Kanto	JPNKN
	Japan/Chubu	JPNCB
	Japan/Kinki	JPNKK
	Japan/Chugoku-Shikoku	JPNCS
	Japan/Kyushu-Okinawa	JPNKO
Other East Asia (OEA)	Korea, Rep of/Whole Country	KORWC
	Korea, DPR/Whole Country	PRKWC
	Mongolia/Whole Country	MNGWC
	Taiwan/Whole Country	TWNWC
Southeast Asia (SEA)	Brunei/Whole Country	BRNWC
	Cambodia/Whole Country	KHMWC
	Indonesia/Whole Country	IDNWC
	Laos/Whole Country	LAOWC
	Malaysia/Whole Country	MYSWC
	Myanmar/Whole Country	MMRW
	Philippines/Whole Country	PHLWC
	Singapore/Whole Country re	SGPWC
	Thailand/Whole Country	THAWC
	Vietnam/Whole Country	VNMWC
Other South Asia (OSA)	Bangladesh/Whole Country	BGDWC
	Bhutan/Whole Country	BTNWC
	Nepal/Whole Country	NPLWC
	Pakistan/Whole Country	PAKWC
	Sri Lanka/Whole Country	LKAWC
	Afghanistan/Whole Country	AFGWC
	Maldives/Whole Country	MDVWC

**Table S2: Uncertainties [%] of emissions of SO<sub>2</sub>, NO<sub>x</sub>, CO, CO<sub>2</sub>, PM<sub>10</sub>, PM<sub>2.5</sub>, BC, and OC for each sector in China, India, Japan, Southeast Asia (SEA), East Asia other than China and Japan (OEA), and South Asia other than India (OSA) in (a) 2015, (b) 1985, and (c) 1955. Abbreviations for sectors are as follows: PP = Power Plants, IND = Industry, ROAD = Road transport, OTRA = Other transport, and DOM = Domestic. For OTRA of OEA in 1955, no emissions were estimated.**

(a) 2015

	SO <sub>2</sub>	NO <sub>x</sub>	CO	CO <sub>2</sub>	PM <sub>10</sub>	PM <sub>2.5</sub>	BC	OC
<b>China</b>								
PP	±38	±65	±78	±48	±83	±83	±83	±83
IND	±48	±70	±113	±40	±115	±116	±160	±153
ROAD	±43	±47	±71	±46	±87	±86	±90	±83
OTRA	±57	±79	±131	±59	±134	±125	±138	±134
DOM	±59	±118	±205	±88	±251	±264	±258	±302
<b>India</b>								
PP	±47	±69	±102	±46	±96	±95	±93	±167
IND	±54	±74	±136	±55	±162	±181	±173	±243
ROAD	±52	±58	±67	±42	±88	±88	±95	±82
OTRA	±70	±84	±121	±66	±112	±112	±112	±112
DOM	±81	±174	±236	±136	±287	±292	±291	±298
<b>Japan</b>								
PP	±36	±43	±89	±41	±87	±89	±90	±93
IND	±43	±59	±64	±39	±96	±99	±89	±108
ROAD	±24	±17	±38	±31	±50	±50	±51	±50
OTRA	±39	±80	±117	±58	±110	±111	±111	±111
DOM	±51	±62	±78	±39	±74	±75	±72	±75
<b>SEA</b>								
PP	±59	±67	±113	±46	±102	±103	±103	±141
IND	±55	±78	±195	±62	±165	±191	±181	±254
ROAD	±54	±65	±109	±46	±92	±91	±89	±97
OTRA	±69	±83	±121	±65	±112	±112	±112	±112
DOM	±95	±173	±219	±137	±286	±288	±289	±291
<b>OEA</b>								
PP	±54	±63	±84	±43	±96	±96	±92	±83
IND	±60	±79	±110	±56	±116	±115	±111	±120
ROAD	±98	±106	±111	±73	±133	±134	±137	±130
OTRA	±58	±89	±126	±71	±117	±117	±117	±117
DOM	±75	±108	±141	±88	±183	±208	±215	±241
<b>OSA</b>								
PP	±64	±49	±84	±43	±78	±80	±76	±106
IND	±63	±87	±207	±65	±146	±163	±189	±251
ROAD	±52	±61	±105	±42	±96	±97	±103	±96
OTRA	±69	±84	±121	±66	±112	±112	±112	±112
DOM	±113	±190	±255	±149	±309	±309	±310	±310

(b) 1985

	SO <sub>2</sub>	NO <sub>x</sub>	CO	CO <sub>2</sub>	PM <sub>10</sub>	PM <sub>2.5</sub>	BC	OC
<b>China</b>								
PP	±50	±95	±82	±51	±93	±93	±83	±92
IND	±56	±113	±135	±52	±120	±109	±165	±154
ROAD	±61	±76	±75	±48	±113	±113	±114	±113
OTRA	±68	±144	±154	±71	±152	±152	±170	±170
DOM	±78	±174	±241	±135	±279	±291	±283	±328
<b>India</b>								
PP	±54	±104	±93	±47	±105	±105	±104	±71
IND	±53	±120	±211	±88	±177	±206	±190	±274
ROAD	±63	±74	±85	±50	±140	±141	±145	±136
OTRA	±67	±92	±86	±53	±139	±117	±110	±98
DOM	±97	±208	±265	±171	±319	±324	±321	±328
<b>Japan</b>								
PP	±41	±78	±74	±37	±83	±83	±65	±73
IND	±45	±82	±62	±42	±96	±102	±107	±124
ROAD	±31	±31	±59	±34	±69	±69	±71	±68
OTRA	±48	±123	±119	±62	±116	±116	±116	±116
DOM	±52	±66	±75	±44	±78	±75	±70	±111
<b>SEA</b>								
PP	±61	±81	±123	±43	±100	±101	±99	±200
IND	±61	±120	±268	±98	±178	±213	±202	±286
ROAD	±62	±79	±111	±52	±115	±115	±119	±113
OTRA	±77	±141	±122	±66	±151	±151	±151	±151
DOM	±114	±210	±237	±166	±316	±317	±318	±319
<b>OEA</b>								
PP	±60	±109	±95	±51	±97	±97	±96	±81
IND	±56	±108	±96	±57	±115	±114	±115	±116
ROAD	±109	±116	±120	±89	±150	±152	±157	±146
OTRA	±75	±126	±126	±71	±116	±116	±116	±116
DOM	±90	±108	±124	±82	±164	±163	±165	±171
<b>OSA</b>								
PP	±63	±69	±80	±39	±90	±92	±80	±76
IND	±57	±114	±240	±86	±176	±205	±214	±272
ROAD	±62	±72	±92	±50	±112	±112	±115	±112
OTRA	±62	±95	±103	±51	±88	±91	±109	±91
DOM	±115	±225	±286	±185	±340	±341	±341	±342

(c) 1955

	SO <sub>2</sub>	NO <sub>x</sub>	CO	CO <sub>2</sub>	PM <sub>10</sub>	PM <sub>2.5</sub>	BC	OC
<b>China</b>								
PP	±126	±140	±129	±105	±131	±131	±124	±131
IND	±124	±160	±176	±101	±127	±121	±188	±166
ROAD	±91	±101	±90	±60	±145	±145	±146	±144
OTRA	±138	±182	±191	±118	±182	±182	±200	±200
DOM	±137	±268	±325	±225	±391	±397	±406	±426
<b>India</b>								
PP	±110	±128	±127	±91	±119	±119	±116	±127
IND	±103	±202	±305	±177	±263	±293	±272	±333
ROAD	±84	±84	±105	±56	±149	±150	±152	±146
OTRA	±139	±145	±138	±109	±185	±174	±170	±158
DOM	±162	±265	±315	±227	±377	±381	±377	±382
<b>Japan</b>								
PP	±109	±123	±124	±92	±144	±145	±135	±130
IND	±97	±125	±62	±88	±110	±114	±151	±128
ROAD	±52	±38	±66	±35	±81	±81	±83	±81
OTRA	±104	±122	±121	±107	±184	±173	±172	±155
DOM	±89	±167	±191	±144	±236	±276	±290	±307
<b>SEA</b>								
PP	±142	±121	±133	±100	±132	±134	±126	±124
IND	±137	±239	±333	±218	±310	±327	±323	±339
ROAD	±89	±91	±131	±69	±136	±136	±138	±134
OTRA	±147	±182	±163	±118	±191	±191	±191	±191
DOM	±183	±266	±282	±221	±369	±369	±369	±370
<b>OEA</b>								
PP	±112	±154	±145	±105	±146	±146	±142	±131
IND	±113	±137	±146	±100	±138	±138	±141	±152
ROAD	±108	±129	±130	±88	±164	±165	±169	±161
OTRA	-	-	-	-	-	-	-	-
DOM	±140	±167	±208	±143	±262	±290	±296	±318
<b>OSA</b>								
PP	±108	±108	±107	±87	±137	±138	±113	±118
IND	±144	±227	±329	±189	±274	±311	±313	±338
ROAD	±89	±78	±90	±64	±111	±111	±115	±111
OTRA	±143	±143	±155	±101	±129	±125	±139	±125
DOM	±180	±282	±336	±242	±394	±394	±394	±394

160 **Table S3: Uncertainties [%] of emissions of NMVOC for each sector in China, India, Japan, Southeast Asia (SEA), East Asia other than China and Japan (OEA), and South Asia other than India (OSA) in 2015, 1985, and 1955.** Abbreviations for sectors are the same as in Table S2 except for EXT = Extraction processes, SLV = Solvent and paint use, and WST = Waste treatment. Note that uncertainties of emissions from non-combustion sources in Japan based on MOEJ (2017) were not assessed in this study. For OTRA of OEA in 1955, no emissions were estimated.

	China	India	Japan	SEA	OEA	OSA
<b>2015</b>						
PP	±75	±110	±90	±120	±80	±86
IND	±101	±111	±39	±136	±246	±144
ROAD	±81	±107	±52	±120	±108	±132
OTRA	±131	±151	±146	±150	±154	±151
DOM	±247	±257	±62	±255	±205	±261
EXT	±130	±125	-	±132	±141	±136
SLV	±150	±149	-	±156	±142	±146
WST	±173	±186	-	±192	±188	±200
<b>1985</b>						
PP	±74	±85	±74	±126	±89	±75
IND	±94	±180	±45	±193	±156	±143
ROAD	±92	±103	±56	±122	±114	±118
OTRA	±154	±117	±149	±151	±154	±117
DOM	±284	±285	±70	±283	±155	±291
EXT	±135	±126	-	±131	±137	±139
SLV	±162	±162	-	±160	±160	±150
WST	±200	±200	-	±200	±200	±200
<b>1955</b>						
PP	±121	±100	±111	±131	±129	±120
IND	±169	±307	±129	±328	±316	±311
ROAD	±110	±102	±60	±154	±124	±117
OTRA	±191	±129	±116	±191	-	±163
DOM	±338	±335	±259	±330	±287	±339
EXT	±178	±168	-	±178	±182	±179
SLV	±162	±149	-	±159	±146	±150
WST	±245	±245	-	±245	±245	±245

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**Table S4: Uncertainties [%] of emissions of NH<sub>3</sub> for each sector in China, India, Japan, Southeast Asia (SEA), East Asia other than China and Japan (OEA), and South Asia other than India (OSA) in 2015, 1985, and 1955.** Abbreviations for sectors are the same as in Table S2 except for MISC = Human (perspiration and respiration) and latrines. Note that uncertainties of emissions from agricultural sources based on REASv1.1 (Yamaji et al., 2004; Yan et al., 2003) were not assessed in this study. For OTRA of OEA in 1955, no emissions were estimated.

	China	India	Japan	SEA	OEA	OSA
<b>2015</b>						
PP	±133	±237	±149	±217	±113	±161
IND	±106	±155	±176	±177	±137	±145
ROAD	±117	±117	±114	±145	±119	±142
OTRA	±163	±170	±164	±170	±174	±170
DOM	±261	±261	±110	±261	±287	±262
MISC	±130	±131	±131	±131	±131	±131
<b>1985</b>						
PP	±140	±128	±157	±276	±133	±134
IND	±99	±187	±202	±224	±138	±151
ROAD	±103	±121	±102	±125	±120	±131
OTRA	±174	±148	±168	±171	±174	±164
DOM	±290	±290	±130	±290	±282	±292
MISC	±132	±132	±132	±132	±132	±132
<b>1955</b>						
PP	±181	±153	±171	±154	±181	±171
IND	±122	±333	±186	±339	±309	±339
ROAD	±125	±154	±116	±180	±133	±160
OTRA	±210	±201	±202	±210	-	±184
DOM	±339	±339	±332	±337	±337	±339
MISC	±133	±133	±133	±133	±133	±133

## References

- Bo, Y., Cai, H., and Xie, S. D.: Spatial and temporal variation of historical anthropogenic NMVOCs emission inventories in China, *Atmos. Chem. Phys.*, 8, 7297–7316, <https://doi.org/10.5194/acp-8-7297-2008>, 2008.
- Fukui, T., Kokuryo, K., Baba, T., and Kannari, A.: Updating EAGrid2000-Japan emissions inventory based on the recent emission trends (in Japanese), *J. Jpn. Soc. Atmos. Environ.*, 2, 117–125, <https://doi.org/10.11298/taiki.49.117>, 2014.
- Garg, A., Shukla, P. R., and Kaphe, M.: The sectoral trends of multigas emissions inventory of India, *Atmos. Environ.*, 40, 4608–4620, <https://doi.org/10.1016/j.atmosenv.2006.03.045>, 2006.
- Huang, X., Song, Y., Li, M., Li, J., Huo, Q., Cai, X., Zhu, T., Hu, M., and Zhang, H.: A high-resolution ammonia emission inventory in China, *Global Biogeochem. Cy.*, 26, GB1030, <https://doi.org/10.1029/2011GB004161>, 2012.
- Kang, Y., Liu, M., Song, Y., Huang, X., Yao, H., Cai, X., Zhang, H., Kang, L., Liu, X., Yan, X., He, H., Zhang, Q., Shao, M., and Zhu, T.: High-resolution ammonia emissions inventories in China from 1980 to 2012, *Atmos. Chem. Phys.*, 16, 2043–2058, <https://doi.org/10.5194/acp-16-2043-2016>, 2016.
- Kannari, A., Baba, T., and Hayami, H.: Estimation of ammonia emissions in Japan (in Japanese), *J. Jpn. Soc. Atmos. Environ.*, 36, 29–38, <https://doi.org/10.11298/taiki1995.36.29>, 2001.
- Kannari, A., Tonooka, Y., Baba, T., and Murano, K.: Development of multiple-species 1 km × 1 km resolution hourly basis emissions inventory for Japan, *Atmos. Environ.*, 41, 3428–3439, <https://doi.org/10.1016/j.atmosenv.2006.12.015>, 2007.
- Kato, N. and Akimoto, H.: Anthropogenic emissions of SO<sub>2</sub> and NO<sub>x</sub> in Asia: emissions inventories, *Atmos. Environ.*, 26, 2997–3017, [https://doi.org/10.1016/0960-1686\(92\)90291-R](https://doi.org/10.1016/0960-1686(92)90291-R), 1992.
- Klimont, Z., Smith, S. J., and Cofala, J.: The last decade of global anthropogenic sulfur dioxide: 2000–2011 emissions, *Environ. Res. Lett.*, 8, 014003, <https://doi.org/10.1088/1748-9326/8/1/014003>, 2013.
- Klimont, Z., Kupiainen, K., Heyes, C., Purohit, P., Cofala, J., Rafaj, P., Borken-Kleefeld, J., and Schöpp, W.: Global anthropogenic emissions of particulate matter including black carbon, *Atmos. Chem. Phys.*, 17, 8681–8723, <https://doi.org/10.5194/acp-17-8681-2017>, 2017.
- Lei, Y., Zhang, Q., He, K. B., and Streets, D. G.: Primary anthropogenic aerosol emission trends for China, 1990–2005, *Atmos. Chem. Phys.*, 11, 931–954, <https://doi.org/10.5194/acp-11-931-2011>, 2011.
- Li, M., Zhang, Q., Zheng, B., Tong, D., Lei, Y., Liu, F., Hong, C., Kang, S., Yan, L., Zhang, Y., Bo, Y., Su, H., Cheng, Y., and He, K.: Persistent growth of anthropogenic non-methane volatile organic compound (NMVOC) emissions in China during 1990–2017: drivers, speciation and ozone formation potential, *Atmos. Chem. Phys.*, 19, 8897–8913, <https://doi.org/10.5194/acp-19-8897-2019>, 2019.
- Lu, Z., Zhang, Q., and Streets, D. G.: Sulfur dioxide and primary carbonaceous aerosol emissions in China and India, 1996–2010, *Atmos. Chem. Phys.*, 11, 9839–9864, <https://doi.org/10.5194/acp-11-9839-2011>, 2011.

- Ma, Q., Cai, S., Wang, S., Zhao, B., Martin, R. V., Brauer, M., Cohen, A., Jiang, J., Zhou, W., Hao, J., Frostad, J.,  
205 Forouzanfar, M. H., and Burnett, R. T.: Impacts of coal burning on ambient PM<sub>2.5</sub> pollution in China, *Atmos. Chem. Phys.*, 17, 4477–4491, <https://doi.org/10.5194/acp-17-4477-2017>, 2017.
- MOEJ (Ministry of Environment of Japan): Report on Volatile Organic Compound (VOC) Emission Inventory Compiled (in Japanese), available at: <http://www.env.go.jp/air/osen/voc/inventory.html> (last access: 23 November 2019), 2017.
- Morth (Ministry of Road Transport & Highways of India): Road Transport Yearbook (2001–2015), Transport Research Wing, Delhi, 2003–2017.
- Pandey, A., Sadavarte, P., Rao, A. B., and Venkataraman, C.: Trends in multi-pollutant emissions from a technology-linked inventory for India: II. Residential, agricultural and informal industry sectors, 99, 341–352, <https://doi.org/10.1016/j.atmosenv.2014.09.080>, 2014.
- Reddy, M. S. and Venkataraman, C.: Inventory of aerosol and sulphur dioxide emissions from India: I – Fossil fuel combustion, *Atmos. Environ.*, 36, 677–697, [https://doi.org/10.1016/S1352-2310\(01\)00463-0](https://doi.org/10.1016/S1352-2310(01)00463-0), 2002a.
- Reddy, M. S. and Venkataraman, C.: Inventory of aerosol and sulphur dioxide emissions from India. Part II – biomass combustion, *Atmos. Environ.*, 36, 699–712, [https://doi.org/10.1016/S1352-2310\(01\)00464-2](https://doi.org/10.1016/S1352-2310(01)00464-2), 2002b.
- Sadavarte, P. and Venkataraman, C.: Trends in multi-pollutant emissions from a technology-linked inventory for India: I. Industry and transport sectors, *Atmos. Environ.*, 99, 353–364, <https://doi.org/10.1016/j.atmosenv.2014.09.081>, 2014.
- 220 Sharma, S., Goel, A., Gupta, D., Kumar, A., Mishra, A., Kundu, S., Chatani, S., and Klimont, Z.: Emission inventory of non-methane volatile organic compounds from anthropogenic sources in India, *Atmos. Environ.*, 102, 209–219, <https://doi.org/10.1016/j.atmosenv.2014.11.070>, 2015.
- Smith, S. J., van Aardenne, J., Klimont, Z., Andres, R. J., Volke, A., and Delgado Arias, S.: Anthropogenic sulfur dioxide emissions: 1850–2005, *Atmos. Chem. Phys.*, 11, 1101–1116, <https://doi.org/10.5194/acp-11-1101-2011>, 2011.
- 225 Streets, D. G., Tsai, N. Y., Akimoto, H., and Oka, K.: Sulfur dioxide emissions in Asia in the period 1985–1997, 34, 4413–4424, [https://doi.org/10.1016/S1352-2310\(00\)00187-4](https://doi.org/10.1016/S1352-2310(00)00187-4), 2000.
- Streets, D. G., Bond, T. C., Carmichael, G. R., Fernandes, S. D., Fu, Q., He, D., Klimont, Z., Nelson, S. M., Tsai, N. Y., Wang, M. Q., Woo, J.-H., and Yarber, K. F.: An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, *J. Geophys. Res.*, 108, 8809, <https://doi.org/10.1029/2002JD003093>, 2003.
- 230 Streets, D. G., Zhang, Q., Wang, L., He, K., Hao, J., Wu, Y., Tang, Y., and Carmichael, G. R.: Revisiting China's CO emissions after the Transport and Chemical Evolution over the Pacific (TRACE-P) mission: Synthesis of inventories, atmospheric modeling, and observations, 111, D14306, <https://doi.org/10.1029/2006JD007118>, 2006.
- Sugiyama, T., Nansai, K., Tohno, S., and Yamamoto, K.: Compilation and application of a primary PM<sub>2.5</sub> emissions inventory with high sectoral resolution in Japan, *Atmos. Environ.*, 43, 759–768, <https://doi.org/10.1016/j.atmosenv.2008.11.003>, 2009.

- Venkataraman, C., Brauer, M., Tibrewal, K., Sadavarte, P., Ma, Q., Cohen, A., Chaliyakunnel, S., Frostad, J., Klimont, Z., Martin, R. V., Millet, D. B., Philip, S., Walker, K., and Wang, S.: Source influence on emission pathways and ambient PM<sub>2.5</sub> pollution over India (2015–2050), *Atmos. Chem. Phys.*, 18, 8017–8039, <https://doi.org/10.5194/acp-18-8017-2018>, 2018.
- Wang, S. X., Zhao, B., Cai, S. Y., Klimont, Z., Nielsen, C. P., Morikawa, T., Woo, J. H., Kim, Y., Fu, X., Xu, J. Y., Hao, J.  
240 M., and He, K. B.: Emission trends and mitigation options for air pollutants in East Asia, *Atmos. Chem. Phys.*, 14, 6571–6603, <https://doi.org/10.5194/acp-14-6571-2014>, 2014.
- Wei, W., Wang, S., Chatani, S., Klimont, Z., Cofala, J., and Hao, J.: Emission and speciation of non-methane volatile organic compounds from anthropogenic sources in China, *Atmos. Environ.*, 42, 4976–4988, <https://doi.org/10.1016/j.atmosenv.2008.02.044>, 2008.
- 245 Xia, Y., Zhao, Y., and Nielsen, C. P.: Benefits of China's efforts in gaseous pollutant control indicated by the bottom-up emissions and satellite observations 2000–2014, 136, 43–53, <https://doi.org/10.1016/j.atmosenv.2016.04.013>, 2016.
- Yamaji, K., Ohara, T., and Akimoto, H.: Regional-specific emission inventory for NH<sub>3</sub>, N<sub>2</sub>O, and CH<sub>4</sub> via animal farming in South, Southeast, and East Asia, *Atmos. Environ.*, 38, 7111–7121, <https://doi.org/10.1016/j.atmosenv.2004.06.045>, 2004.
- Yan, X., Akimoto, H., and Ohara, T.: Estimation of nitrous oxide, nitric oxide, and ammonia emissions from croplands in  
250 East, Southeast, and South Asia, *Global Change Biol.*, 9, 1080–1096, <https://doi.org/10.1046/j.1365-2486.2003.00649.x>, 2003.
- Zhang, Q., Streets, D. G., Carmichael, G. R., He, K. B., Huo, H., Kannari, A., Klimont, Z., Park, I. S., Reddy, S., Fu, J. S., Chen, D., Duan, L., Lei, Y., Wang, L. T., and Yao, Z. L.: Asian emissions in 2006 for the NASA INTEX-B mission, *Atmos. Chem. Phys.*, 9, 5131–5153, <https://doi.org/10.5194/acp-9-5131-2009>, 2009.
- 255 Zhao, Y., Nielsen, C. P., Lei, Y., McElroy, M. B., and Hao, J.: Quantifying the uncertainties of a bottom-up emission inventory of anthropogenic atmospheric pollutants in China, *Atmos. Chem. Phys.*, 11, 2295–2308, <https://doi.org/10.5194/acp-11-2295-2011>, 2011.
- Zhao, Y., Nielsen, C. P., McElroy, M. B., Zhang, L., and Zhang, J.: CO emissions in China: uncertainties and implications of improved energy efficiency and emission control, *Atmos. Environ.*, 49, 103–113,  
260 <https://doi.org/10.1016/j.atmosenv.2011.12.015>, 2012.
- Zhao, Y., Zhang, J., and Nielsen, C. P.: The effects of recent control policies on trends in emissions of anthropogenic atmospheric pollutants and CO<sub>2</sub> in China, *Atmos. Chem. Phys.*, 13, 487–508, <https://doi.org/10.5194/acp-13-487-2013>, 2013.
- Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L., Qi, J., Yan, L., Zhang, Y., Zhao, H., Zheng, Y., He, K., and Zhang, Q.: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions,  
265 *Atmos. Chem. Phys.*, 18, 14095–14111, <https://doi.org/10.5194/acp-18-14095-2018>, 2018.