Supporting Information

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SI Text

SI Methods

We applied the NASA Goddard Institute for Space Studies (GISS) atmospheric composition-climate model described in detail and comprehensively evaluated in (22). Briefly, the model comprises the GISS version ModelE general circulation model (35) with embedded fully interactive photochemistry and aerosol modules. We use 23 vertical layers (model top in the mesosphere) and 4×5 degree latitudinal by longitudinal horizontal resolution. The atmospheric composition-climate model includes full coupling between tropospheric gas-phase and aerosol chemistry (36, 37) and aerosols and cloud microphysics for liquid-phase stratus and cumulus clouds (38). The aerosols are assumed to be externally mixed. The direct instantaneous TOA RF by the SLS is calculated internally within the climate model's radiation scheme (35). We estimate the RF due to AIE from the difference between the net radiation at the TOA and all direct radiative effects (38).

Simulations. In all simulations using the atmospheric composition-climate model, monthly mean sea surface temperatures and sea ice climatologies are prescribed for 1990–1999 (1). Methane concentration is prescribed to hemispherically averaged values ($\mathrm{NH} = 1,814$ ppb and $\mathrm{SH} = 1,733$ ppb) based on observations for the year 2000 (2). A present day control simulation is performed based on the emissions inventory described in the paper and Table S1. In order to quantify the short-lived species (SLS) radiative forcing (RF) attributable to each emission sector, we performed sensitivity simulations in which we removed all emissions from that sector. Each simulation was run for 12 model years; the first 2 years of the simulations are discarded as spin-up and the remaining 10 years are averaged. The contributions to RF by the individual emissions sectors for each SLS are then determined by taking the difference between the control simulation and the simulation with a missing sector. A further simulation was performed in which all sectors were removed simultaneously.

CH_4 RF Due to Indirect Chemical Effects on the CH_4 Lifetime. Indirect CH_4 RF is determined using the method described in ref. 3. First the change in atmospheric CH_4 concentration due to each emission sector is calculated based on the initial change in the CH_4 lifetime in the climate model and accounting for the feedback of CH_4 on its own lifetime. Then the CH_4 RF is calculated using a standard simplified expression based on the steady-state concentration change (4). Both direct and indirect CH_4 changes also affect O_3 on the longer time scale of the CH_4 lifetime. We quantify this secondary O_3 RF (denoted "M-O_3") using global average sensitivity results from previous multimodel assessment studies (3, 4).

Aerosol Indirect Effect. Aerosol mass concentrations are converted to aerosol number concentrations assuming log-normal distributions and are related to cloud droplet number concentrations (CDNC) through empirical equations. Additionally, effects of changes to cloud cover and turbulence on CDNC are also included. To represent aerosol effects on precipitation, the autoconversion scheme in the model is modified to include a dependence on droplet size (as well as CDNC and droplet dispersion effects) such that autoconversion is triggered if droplet sizes exceed 14 μm (5). Model simulated aerosol-cloud interactions been evaluated with satellite-based retrievals to constrain the magnitude of the aerosol indirect effect (AIE) (6). Results indicate that the AIE may be overpredicted over the ocean regions due to an underprediction of cloud droplet size and an overprediction of cloud optical depth. Simulated CDNC was found to be within satellite retrieved uncertainty. Over land locations, coincident retrievals of aerosols and cloud properties are not easily available from satellites and thus evaluation of changes to cloud properties from aerosols is more difficult. A more meaningful global evaluation of the AIE is challenging in the absence of global observations of CDNC that serve as the main link between aerosols and cloud properties.

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Table S1. Global anthropogenic emissions inventory based on Edgar Fast Track 2000 (7, 8); units for CO, NMVOC, SO_2 CH_4, N_2O, and CO_2 are Teragram (Tg)Full Molecular Mass (FMM)/year; units for NO_x and NH_3 are TgNitrogen(N)/year; units for black carbon and organic carbon are Gigagram(GgFMM/year

	Precursor PrePspecies									
Sector	NO_x	CO	NMVOC	SO_2	BC	OC	CH_4	NH_3	N_2O	CO_2
Industry	6.0	51	33.6	63.2	769	2559	2.7	0.2	0.7	8414
Power	7.8	12	33.3	57.7	22	18	93.9	0.1	0.1	9127
Household fossil fuel	0.9	27	1.2	8.1	453	486	1.7	2.2	0.02	3390
Household biofuel	2.2	237	27.3	3.1	1471	7823	13.8	0	0.2	495
On-road transportation	8.7	186	33.8	3.7	1235	1630	0.9	0	0.1	4276
Off-road (land) transportation	1.8	13	4.6	2.0	588	292	0.008	0	0.003	390
Shipping	2.9	0.1	0.02	7.3	97	136	0.028	0	0.003	428
Aviation	0.7	0	0	0.2	11	0	0.006	0	0.020	654
Agricultural waste burning	0.2	16	2.0	0.2	371	2266	0.8	1.4	0.020	0
Waste/landfill	0.04	4	2.7	0.05	0	0	58.2	2.7	0.3	0
Biomass burning	10.2	507	31.3	2.7	3500	37200	21.2	1.8	0.9	2740
Animals	0	0	0	0	0	0	88.5	21.1	3.2	0
Agriculture	0	0	0	0	0	0	39.4	12.6	6.6	0

Table S2. Global annual average direct RF due to SLS and AIE RF by sector

	Ozone, S-O_3	Sulfate	Nitrate	Black carbon	Organic carbon	AIE
Industry	14* (3)	−192* (3)	47* (1)	49* (1)	−15* (3)	−275* (49)
Power	7* (2)	−164* (3)	25* (2)	4 (1)	4 (3)	−182* (46)
Biomass burning	60* (3)	−11* (2)	−21* (2)	131* (1)	−203* (2)	−179* (50)
Agriculture	−7* (3)	1 (3)	−59* (2)	−1 (1)	1 (2)	——
Aviation	3 (3)	−17* (3)	−3* (1)	3 (2)	1 (3)	——
Agr. waste burning	1 (2)	−11* (3)	−11* (2)	21* (4)	−19* (2)	——
Household fossil fuel	2 (2)	−26* (5)	−6* (2)	25* (1)	−5* (2)	——
Shipping	3 (2)	−16* (5)	−16* (2)	3 (2)	−3 (2)	——
Off-road land	2 (2)	−10* (4)	−7* (2)	33* (2)	−2 (3)	——
On-road	26* (2)	−14* (2)	−25* (2)	71* (2)	−16* (2)	43* (33)
Household biofuel	21* (2)	−7 (4)	−4* (2)	82* (1)	−64* (2)	32* (30)
Animals	−4 (3)	5 (3)	−83* (2)	−2 (1)	3 (3)	——
Waste/landfill	−2 (2)	−6 (3)	−16* (2)	1 (1)	−1 (3)	——

Values marked with * indicate significance at 95% confidence level and bracketed numbers indicate standard error relative to internal climate variability based on 10 years of model output. AIE RF values are shown only for results with significance at 95% confidence level. Units are mWm^{-2}.

Table S3. Global annual average RF from methane direct effects (D-CH_4) and indirect chemical effects of SLS precursors on CH_4 lifetime (I-CH_4) and associated secondary ozone RF (M-O_3) (mWm^{-2})

	Direct Methane Emission		Indirect chemical Effect	
	D-CH_4	M-O_3	I-CH_4	M-O_3
Industry	4	1	−9	−4
Power	128	46	−21	−9
Biomass burning	28	11	6	2
Agriculture	53	19	0.4	0.2
Aviation	0	0	−6	−3
Agr. waste burning	1	0.4	3	1
Household fossil fuel	2	0.8	2	1
Shipping	0.1	0.01	−18	−7
Off-road land	0	0	−4	−2
On-road	1.2	0.4	1.3	0.6
Household biofuel	18	7	24	10
Animals	120	43	2	1
Waste/landfill	78	29	−0.5	0.2
Sum	433	158	−20	−9

Table S4. Global annual average RF by long-lived greenhouse gases due to perpetual constant year 2000 emissions by sector calculated using the MAGICC model at future time points 2020 and 2100 relative to 2000 (mWm^{-2})

	Future impact of year 2000 emissions			
	2020		2100	
	N_2O	CO_2	N_2O	CO_2
Industry	3	219	10	653
Power	0.4	240	1.4	714
Biomass burning	3	68	12	186
Agriculture	26	0	95	0
Aviation	0	16	0	49
Agr. waste burning	0	0	0	0
Household fossil fuel	0	88	0	258
Shipping	0	11	0	32
Off-road land	0	10	0	29
On-road	0.4	110	2	326
Household biofuel	0.6	13	2	38
Animals	12	0	45	0
Waste/landfill	1	0.4	4	2
Sum	46	775	171	2287

Table S5. Uncertainties in sectoral RFs. Total RF by sector at 2020 and 2100 and the ratio of the SLS/LLGHG RF at each time point (SLS includes CH_4). RF uncertainty is given in terms of internal variability in the climate model based on the standard error of the mean for 10 years of model output, and in terms of the emission rate for the SLS, in both cases as a percentage of the total sector RF. Sectors that emit species with opposing RF and high uncertainties are indicated using '*'.

SECTOR	RF at 2020	SLS/LLGHG at 2020	RF at 2100	SLS/LLGHG at 2100	Uncertainty due to internal climate variability at 2020	Uncertainty due to emission rate at 2020	Large opposing uncertainties
Power	79	-0.7	554	-0.2	70%	50%	
On-road transportation	199	0.8	417	0.3	20%	40%	
Household fossil fuel	84	-0.05	254	-0.01	10%	10%	*
Household biofuel	132	8.7	159	3	30%	160%	*
Animal husbandry	98	7.2	131	1.9	10%	90%	
Industry	-158	-1.7	283	-0.6	40%	120%	*
Agriculture	29	0.1	98	0.03	40%	10%	
Waste/Landfill	84	58.9	88	13.7	10%	100%	
Off-road (land) transportation	20	1	39	0.3	70%	60%	*
Aviation	-6	-1.4	27	-0.4	200%	180%	
Agriculture waste burning	-14	N/A	-14	N/A	90%	250%	*
Biomass burning	-106	-2.5	22	-0.9	60%	420%	*
Shipping	-43	-4.9	-22	-1.7	30%	130%	*

Table S6. Comparison of total RF by species summed over all sectors to relevant Intergovernmental Panel on Climate Change (IPCC) AR4 values

Component	IPCC AR4	This study
CH_4	0.48 [± 0.05]	+0.41
Total tropospheric O_3	+0.35 [−0.1,+0.3]	+0.42
Tropospheric O_3 from CH_4	+0.2	+0.15
Total direct aerosol	−0.50 [± 0.4]	−0.55
Direct sulfate aerosol	−0.40 [±0.20]	−0.47
Direct fossil fuel organic carbon	−0.05 [±0.05]	−0.04
Direct fossil fuel black carbon	+0.2 [±0.15]	+0.19
Direct biomass burning aerosol	+0.03 [±0.12]	−0.10
Direct nitrate	−0.10 [±0.10]	−0.17
AIE	−0.70 [−1.1,+0.4]	−0.57

IPCC AIE includes cloud albedo affect only, whereas this study includes cloud albedo, cloud lifetime, and semidirect effects.