

## **Supporting Information**

# **The enhancement of local air pollution by urban CO<sub>2</sub> domes**

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### **Summary**

This Supporting Information describes the model and emissions used for this study (Section S1), feedbacks in the model (Section S2), and a description of simulations (Section S3).

## **S1. Description of the Model**

The model used for this study was GATOR-GCMOM, a one-way-nested global-regional Gas, Aerosol, Transport, Radiation, General Circulation, Mesoscale, and Ocean Model that simulates climate, weather, and air pollution on multiple scales [Jacobson, 2001; 2006, 2008; Jacobson *et al.*, 2007, 2009]. The model treated time-dependent gas, aerosol, cloud, radiative, dynamical, ground surface, ocean, and transport processes. It solved 2-D ocean dynamics with a potential-ensrophy, vorticity-, energy-, and mass-conserving scheme [Ketefian and Jacobson, 2009], 3-D ocean energy and chemical diffusion among all gases, including CO<sub>2</sub>, 3-D ocean equilibrium chemistry among >50 reactions in the Na-Cl-Mg-Ca-K-H-O-Li-Sr-C-S-N-Br-F-B-Si-P system each time step, and ocean-atmosphere exchange for all gases [Jacobson, 2005d]. It also treated soil moisture and energy transport among 10 subsurface layers for each of up to 13 subgrid soil types and through roads, rooftops, vegetation, and snow over soil [Jacobson, 2001]. It solved for depths and temperatures of slab sea ice and snow over sea ice, accounting for energy fluxes below ice, between ice and snow, and above ice or snow [Jacobson, 2001], and it predicted rather than prescribed the albedos of snow and sea ice [Jacobson, 2004].

### S1.A. CO<sub>2</sub> and Other Gas Processes

Gas processes included emissions, photochemistry, gas-to-aerosol conversion, gas-to-cloud conversion, gas-cloud chemical exchange, gas-ocean chemical and moisture exchange, advection, convection, molecular diffusion, turbulent diffusion, and dry deposition. Gas chemistry was solved with SMVGEAR II among 134 gases and 347 tropospheric and stratospheric kinetic, heterogeneous, and photolysis reactions [listed in Jacobson, 2008]. Photolysis and UV/visible heating rates were solved explicitly over 86 solar wavelength intervals for each photolyzing and UV- and or visible-absorbing gas. All photolyzing gases fed back to radiative heating rates. With respect to CO<sub>2</sub>, the model treated chlorophyll photosynthesis in ocean and lake water, green-plant photosynthesis over land, chlorophyll and leaf respiration, soil respiration, CO<sub>2</sub> dissolution in/evaporation from clouds, precipitation, lakes, rivers, and oceans, weathering of silicate rocks, natural emissions, anthropogenic emissions, chemical production by atmospheric gases, ocean equilibrium chemistry, ocean formation of carbonate minerals, and CO<sub>2</sub> diffusive transport to the deep [Jacobson, 2005d, Jacobson and Streets, 2009].

### S1.B. Aerosol Processes

Aerosol processes included anthropogenic and natural emissions, binary and ternary homogeneous nucleation, condensation, dissolution, internal-particle chemical equilibrium, aerosol-aerosol coagulation, aerosol-hydrometeor coagulation, sedimentation, dry deposition, advection, convection, molecular diffusion, and turbulent diffusion [Jacobson, 2002a; Jacobson *et al.*, 2007; 2009]. The model treated one discrete aerosol size distribution with 14 size bins (2 nm to 50 μm in diameter), and three hydrometeor distributions, each with 30 size bins (0.5 μm to 8 mm in diameter) (Table S1). Particle number and mole concentrations of several chemicals were tracked in each aerosol and hydrometeor size bin of each size distribution (Table S1). The components within each bin of each distribution were internally mixed in the bin but externally mixed from other bins and other distributions.

**Table S1.** Aerosol and hydrometeor discrete size distributions treated in the model and the parameters (number concentration and chemical mole concentrations) present in each size bin of each distribution. The aerosol distribution contained 14 size bins each, and the hydrometeor distributions contained 30 size bins each. The components within each size bin of each size distribution were internally mixed in the bin but externally mixed from other bins and other distributions.

Aerosol Internally Mixed (IM)	Cloud / Precipitation Liquid	Cloud / Precipitation Ice	Cloud / Precipitation Graupel
Number	Number	Number	Number
BC	BC	BC	BC
POM	POM	POM	POM
SOM	SOM	SOM	SOM
H <sub>2</sub> O(aq)-h	H <sub>2</sub> O(aq)-h	H <sub>2</sub> O(aq)-h	H <sub>2</sub> O(aq)-h
H <sub>2</sub> SO <sub>4</sub> (aq)	H <sub>2</sub> SO <sub>4</sub> (aq)	H <sub>2</sub> SO <sub>4</sub> (aq)	H <sub>2</sub> SO <sub>4</sub> (aq)
HSO <sub>4</sub> <sup>-</sup>	HSO <sub>4</sub> <sup>-</sup>	HSO <sub>4</sub> <sup>-</sup>	HSO <sub>4</sub> <sup>-</sup>
SO <sub>4</sub> <sup>2-</sup>	SO <sub>4</sub> <sup>2-</sup>	SO <sub>4</sub> <sup>2-</sup>	SO <sub>4</sub> <sup>2-</sup>
NO <sub>3</sub> <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>
Cl <sup>-</sup>	Cl <sup>-</sup>	Cl <sup>-</sup>	Cl <sup>-</sup>
H <sup>+</sup>	H <sup>+</sup>	H <sup>+</sup>	H <sup>+</sup>
NH <sub>4</sub> <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>
NH <sub>4</sub> NO <sub>3</sub> (s)	NH <sub>4</sub> NO <sub>3</sub> (s)	NH <sub>4</sub> NO <sub>3</sub> (s)	NH <sub>4</sub> NO <sub>3</sub> (s)
(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> (s)	(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> (s)	(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> (s)	(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> (s)
Na <sup>+</sup> (K, Mg, Ca)	Na <sup>+</sup> (K, Mg, Ca)	Na <sup>+</sup> (K, Mg, Ca)	Na <sup>+</sup> (K, Mg, Ca)
Soil dust	Soil dust	Soil dust	Soil dust
Poll/spores/bact	Poll/spores/bact	Poll/spores/bact	Poll/spores/bact
	H <sub>2</sub> O(aq)-c	H <sub>2</sub> O(s)	H <sub>2</sub> O(s)

POM is primary organic matter; SOM is secondary organic matter. H<sub>2</sub>O(aq)-h is liquid water hydrated onto dissolved ions and undissociated molecules in solution. H<sub>2</sub>O(aq)-c is water that condensed to form liquid hydrometeors, and S(VI)= H<sub>2</sub>SO<sub>4</sub>(aq) + HSO<sub>4</sub><sup>-</sup>+ SO<sub>4</sub><sup>2-</sup>. Condensed and hydrated water existed in the same particles so that, if condensed water evaporated, the core material, including its hydrated water, remained. H<sub>2</sub>O(s) was either water that froze or deposited from the gas phase as ice. The emitted aerosol species included BC, POM, H<sub>2</sub>SO<sub>4</sub>(aq), HSO<sub>4</sub><sup>-</sup>, and SO<sub>4</sub><sup>2-</sup> for fossil-fuel soot; H<sub>2</sub>O, Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>, Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, H<sub>2</sub>SO<sub>4</sub>(aq), HSO<sub>4</sub><sup>-</sup>, and SO<sub>4</sub><sup>2-</sup> for sea spray; the same plus BC and POM for biomass and biofuel burning; soil dust, and pollen/spores/bacteria. In all cases, K<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup> were treated as equivalent Na<sup>+</sup>. Soil dust was generic. Homogeneously nucleated aerosol components included H<sub>2</sub>O, H<sub>2</sub>SO<sub>4</sub>(aq), HSO<sub>4</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup>. Condensing gases included H<sub>2</sub>SO<sub>4</sub> and SOM. Dissolving gases included HNO<sub>3</sub>, HCl, and NH<sub>3</sub>. The liquid water content and H<sup>+</sup> in each bin were determined as a function of the relative humidity and ion composition from equilibrium calculations. All aerosol and hydrometeor distributions were affected by self-coagulation loss to larger sizes and heterocoagulation loss to other distributions (except the graupel distribution, which had no heterocoagulation loss).

Particle sources included vehicles, power plants, industry, ships, aircraft, the ocean (sea spray, bacteria), soils (dust, bacteria), volcanos, vegetation (pollen, spores), biofuel burning, and biomass burning. The baseline anthropogenic emission inventory used for each non-global domain was the U.S. National Emission Inventory (NEI) for 2002 [USEPA, 2006]. Table S2 shows the emission rates in the annual average from the inventory. Global-domain anthropogenic and natural emissions are summarized in *Jacobson and Streets* [2009]. Natural emissions in each regional domain were calculated with the same techniques as in the global domain.

**Table S2.** Fossil-Fuel Emission Rates of Gases and Particles in the Non-Global Domains of the Simulations. Data from USEPA [2006]. Biofuel and biomass-burning emissions were calculated separately.

Species	2002 Los Angeles Basin (Gg/yr)	2002 California/ Nevada (Gg/yr)	2002 U.S. (Gg/yr)

Carbon monoxide	2540	5200	94,100
Carbon dioxide	253,000	460,000	5,762,000
Nitrogen oxides as NO <sub>2</sub>	607	1130	19,500
Organic gases			
Methane	299	729	4380
Ethane	33.2	101	441
Propane	8.95	15.1	226
Paraffin bond group	262	500	8190
Ethene	23.6	45.9	732
Propene	4.93	9.75	153
1,3-Butadiene	4.68	8.81	128
Olefin bond group	5.77	12.0	257
Methanol	0.67	1.23	30.0
Ethanol	6.34	17.7	309
Formaldehyde	6.07	12.0	157
Acetaldehyde	1.80	3.48	45.8
Higher aldehydes	15.3	33.6	525
Formic acid	0.18	0.33	6.4
Acetic acid	0.31	0.57	12.7
Acetone	4.31	12.1	109
Benzene	7.03	13.9	244
Toluene bond group	46.4	86.3	1790
Xylene bond group	46.0	99.6	2680
Isoprene bond group	0.34	0.65	11.0
Unreactive	48.5	91.1	1650
Total organic gas	825	1800	2210
Sulfur oxides as SO <sub>2</sub>	46.6	107	15,860
Ammonia	75.9	98	4530
PM <sub>2.5</sub>			
Organic matter	72.6	188	1710
Black carbon	16.3	38.9	413
Sulfate	4.78	10.6	151
Nitrate	0.474	1.36	16.2
Other	98.0	263	7300
Total PM <sub>2.5</sub>	192	502	9590
PM <sub>10</sub>			
Organic matter	142	354	4020
Black carbon	29.0	68.6	661
Sulfate	8.18	18.7	256
Nitrate	1.10	3.0	53.3
Other	388	982	34,300
Total PM <sub>10</sub>	568	1430	39,300

The mass concentration of each chemical emitted into the aerosol size distribution of each grid cell was first fit to a continuous multi-modal lognormal distribution. For emitted fossil-fuel soot, the distribution was similar to that in *Jacobson et al.* [2005]. For biofuel and biomass-burning particles, the distribution was similar to that for smoldering burn in *Reid and Hobbs* [1998]. The continuous distributions were then discretized into individual model size bins. Black carbon (BC) was assumed to consist of an aggregate of spherules with the smallest spherule diameter of 14 nm [*Jacobson et al.*, 2005]. Since black carbon was emitted within soot particles (BC aggregates coated by POM and S(VI)), the smallest bin that BC could enter was set to 23.8 nm to ensure that if POM and S(VI) evaporated, the BC core would be no smaller than 14 nm. Fossil-fuel POM (which

consists primarily of lubricating oil and unburned fuel oil) and S(VI) existed down to 0.8-nm diameter, the low diameter of the smallest bin. Thus, below 23.8 nm, emitted POM and S(VI) were well-mixed in the same particles. Above this size, emitted BC, POM, and S(VI) were well-mixed in such particles, but with BC treated as a core for optical calculations. Liquid water could then hydrate to the S(VI) and POM [which was less soluble than S(VI)] in the particles, and  $\text{NH}_3$ ,  $\text{HNO}_3$ , and HCl could dissolve/evaporate (through nonequilibrium growth equations) in the water and dissociate/crystallize (through internal-aerosol equilibrium equations), forming ions or the solids,  $\text{NH}_4\text{NO}_3$  and  $(\text{NH}_4)_2\text{SO}_4$  [Jacobson, 2005b].

Homogenous nucleation of sulfuric acid-water-ammonium produced new particles and was solved simultaneously with condensation of sulfuric acid onto the aerosol distribution in order to allow competition of sulfuric acid vapor between nucleation and condensation [Jacobson, 2002a]. Aerosol-aerosol coagulation was solved with a volume- and volume-concentration conserving, noniterative, positive-definite, and unconditionally stable coagulation scheme [Jacobson, 2002a]. The total collision kernel for aerosol coagulation was the combination of kernels for Brownian motion, Brownian diffusion enhancement, van der Waal's forces, viscous forces, fractal geometry, gravitational settling, turbulent shear, and turbulent inertial motion [e.g., Jacobson, 2005a].

#### S1.C. Convective Cloud, Stratus Cloud, Aerosol-Cloud Processes

On both regional and global scales, the model treated first and second indirect effects explicitly and together with cloud radiative effects (Section S1.D). On regional scales, the model treated cloud thermodynamics, size- and composition-resolved microphysics, and transport explicitly. On the global scale and coarsely-resolved regional scales (e.g., on the U.S. domain in this application), the model treated cloud microphysics and vertical transport explicitly but parameterized thermodynamics and did not treat horizontal cloud transport.

The explicit treatment of all cloud types on regional scales (e.g., California, Los Angeles in this application) is described in detail in Jacobson *et al.* [2007].

On the global and coarse regional scales, the model treated subgrid cumulus and grid-scale stratiform clouds. Cumulus thermodynamics were calculated with an Arakawa-Schubert parameterization that treated 0-500 subgrid cumulus clouds in each column, each with a different cloud base and top [Ding and Randall, 1998]. Gases and size- and composition-resolved aerosol particles were convected within each subgrid cumulus cloud [Jacobson, 2003, Section 3.2], and formation/dissipation of each cloud fed back to grid-scale temperature and moisture. Column cumulus cloud fractions were calculated by summing cloud fractions among all subgrid cumulus (no overlap), where each subgrid fraction was determined as the maximum ratio, among all layers in the column, of cloud to air mass in the layer [Jacobson, 2003].

Grid-scale stratiform (including cirrus) liquid and ice cloud fractions and water on the global and coarse-regional scales were calculated by integrating a vertical bivariate normal distribution for energy and moisture determined from their gradients, affected by mechanical and thermal turbulence parameters (representing subgrid eddies) in all model levels, as in Mellor and Yamada [MY, 1982]. Turbulence parameters for the calculation were determined from the MY level 2.5 closure scheme. Since cumulus and stratus cloud fractions were calculated from thermodynamic and moisture variables, size-and-composition-resolved ice microphysics affected cloud fractions only by growing/shrinking, thereby affecting moisture and thermodynamic fields. However, other cloud optical properties (e.g., optical depth, liquid and ice contents, precipitation,

scavenging, etc.) were calculated explicitly from hydrometeor particle shape/size/composition, as described next.

Following subgrid cumulus and stratiform cloud thermodynamics, which provided a quasi-equilibrium estimate of cloud liquid, ice, precipitation, gases, and aerosol particles (as a function of size distribution, size, and composition) in each vertical layer of each subgrid cloud, the cloud water was evaporated/sublimated then regrown onto the discrete size-resolved aerosols in a time-dependent manner, as described in *Jacobson* [2003]. Condensational and depositional growth/evaporation of water was solved simultaneously on both the EFFS and IM aerosol distributions and simultaneously for liquid, ice, or both. The numerical growth scheme was unconditionally stable and mass conservative. The saturation ratios at equilibrium for the growth ordinary differential equations were calculated from Köhler theory assuming the Kelvin effect and Raoult's law affected the equilibrium saturation ratio over liquid water whereas only the Kelvin effect affected that over ice. Whether aerosol particles in a given size bin of a size distribution could activate into cloud drops depended on whether (a) their size and the ambient saturation ratio exceeded the critical size and saturation ratio at equilibrium, as determined by the Köhler equation and (b) the growth equations predicted growth to occur over a particular bin when competition among the gas phase and water in all potentially activatable aerosol particles among all distributions was accounted for [*Jacobson, 2003*].

Following (1) growth, the following size-resolved microphysical calculations were performed: (2) diffusiophoretic, thermophoretic, gravitation, Brownian, etc. coagulation among liquid ice, and graupel and their aerosol components, (3) contact freezing of drops by size-resolved interstitial aerosols, (4) heterogeneous plus homogeneous freezing of liquid drops, (5) liquid drop breakup, (6) coagulation of cloud hydrometeors and incorporated aerosols with interstitial aerosols, (7) sedimentation of liquid, ice, and graupel hydrometeor particles and their aerosol inclusions, (8) coagulation of precipitation hydrometeors with interstitial and below-cloud aerosols (washout), (9) removal of precipitation and incorporated aerosols (rainout), (10) below-cloud evaporation/sublimation to smaller hydrometeors and aerosol cores, (11) gas washout, (12) aqueous chemistry within liquid cloud and aerosol particles, (13) heterogeneous chemistry on ice crystals [*Jacobson, 2008a*], and (14) lightning production due to size-resolved hydrometeor bounceoffs [*Jacobson and Streets, 2009*]. These processes were discrete size- and composition-resolved and accounted for the tracking of all aerosol chemical components within all cloud particles of all sizes.

Aerosol-hydrometeor coagulation interactions occurred among each the EFFS and IM aerosol distributions and the liquid, ice, and graupel hydrometeor distributions (Table S1). Such interactions resulted in loss of aerosol number and component mass from the EFFS and IM distributions and gains to hydrometeor size bins. When liquid, ice, or graupel hydrometeor particles in a size bin evaporated or sublimated, they released their aerosol core number and component mass back to the aerosol size bin corresponding to the single-particle volume of the aerosol-inclusion in the hydrometeor bin, calculated from the core component mass, number, and density. Thus, if only cloud drop growth and evaporation were theoretically considered (e.g., if coagulation, aqueous chemistry, etc. were ignored) and particles grew to cloud drops, then evaporated, the original aerosol distribution would be restored exactly.

Ice crystals formed in one of four time-dependent ways: heterogeneous nucleation followed by ice growth, contact freezing of liquid drops, homogeneous plus heterogeneous freezing of liquid drops, and evaporative freezing of liquid drops [*Jacobson, 2003*]. All methods were discretely size resolved.

Aerosol number concentrations were multiplied by their single-particle cross-sectional areas and single-particle absorption, scattering, and forward scattering efficiencies as a function of wavelength to determine spectral absorption and scattering optical depths and asymmetry parameters, respectively, for each aerosol size distribution. Mie calculations of efficiencies for each particle size and solar plus thermal-IR wavelength (Section S1.D) accounted for absorption by BC inclusions (a core surrounded by a shell of lubricating oil, sulfate and, for age particles, condensed or coagulated material), where the BC concentration in each particle size bin was tracked during the microphysical calculations [Jacobson, 2006]. Overall grid-scale cloud fraction was determined by assuming random overlap among all subgrid cumulus clouds, and grid-scale stratiform clouds in the column

#### S1.D. Radiative Processes

Radiative transfer was solved in the model to determine actinic fluxes for photolysis calculations and irradiances for heating rate calculations. Radiative transfer was solved separately over cloudy and clear portions of each model column, and the results were weighted by clear and cloudy-sky fractions. The radiative solutions for both actinic flux and irradiance [Toon *et al.*, 1988] were found for each of 694 wavelengths/probability intervals in each column (including 86 below 800 nm), with gas (including CO<sub>2</sub>, H<sub>2</sub>O, CH<sub>4</sub>, N<sub>2</sub>O, O<sub>2</sub>, O<sub>3</sub>, CFC) absorption coefficients as in Jacobson [2005c], and aerosol and cloud optical properties as in Jacobson [2002a, 2006]. Ice crystals were treated as nonspherical collections of spheres with the same total volume-to-area ratio as the nonspherical particle [Jacobson, 2006]. Radiative transfer was solved through both the air and a single layer of snow, sea ice, or water, where they existed, so albedos over these surface were calculated, not prescribed. Since the model tracked soot and soil dust inclusions within precipitation, which fell on snow and sea ice, radiative transfer accounted for the optics of soot and soil dust in snow and sea ice [Jacobson, 2004] as well as in individual aerosol particles and cloud drops. In aerosol particles, black carbon was treated as a core surrounded by a mixed shell [Ackerman and Toon, 1981]. In individual sizes of cloud drops, ice crystals, and graupel particles, BC aggregates were treated as polydisperse spherules whose optical properties were calculated with the iterative dynamic effective medium approximation (DEMA) of Chylek *et al.* [1984]. A comparison of this treatment with the core-shell treatment for global black carbon radiative effects is given in Jacobson [2006].

#### **S2. Feedbacks in the Model**

The model treated feedbacks among meteorological, gas, aerosol, cloud, radiative, land surface, and ocean surface process in all nested domains. Since meteorology was coupled online to the other processes, including clouds, photochemistry, aerosol processes, the oceans, etc., any process that affected meteorology fed back to the other processes and vice versa. The main coupling between gas, aerosol, and cloud processes and meteorology was through radiative transfer. For example, radiative heating rates, used to determine changes in temperature, thus changes in pressure, were calculated by accounting for the optical properties of gases, aerosol particles, and hydrometeor particles and their inclusions. A second coupling was through water vapor. Changes in water vapor due to cloud or aerosol growth/evaporation or through air-ocean or air-groundwater exchange, changed air pressure (since water vapor is a component of air pressure in the model) and the specific heat of air. Conversely, changes in meteorological variables, such as air pressure, wind speed/direction, temperature, and the relative humidity, affected the movement and concentrations of gas, aerosol particles, and hydrometeor particles.

At a deeper level, gases in the model were both affected by and affected photolysis and radiative heating rates. All gases that photolyzed in the model affected radiative heating rates since absorption cross sections of each gas at each wavelength were used

both for actinic flux calculations for photolysis (when quantum yields were applied) and irradiance calculations (for heating rates). Thus, when gas concentrations changed, so did photolysis and heating rates. Likewise, aerosol particles, cloud drops, ice crystals, and graupel particles affected gas photolysis and radiative heating rates since particle absorption cross sections were determined as a function of wavelength and included in optical depths used in actinic flux and irradiance calculations. Changes in radiative heating rates in turn affected temperatures, which affected the rates of gas and aerosol chemical reactions, water fluxes from the ocean, the relative humidity, air pressure, wind speeds, cloud and aerosol microphysical processes, etc.

Aerosol particles fed back to clouds and clouds, in turn, fed back to aerosol particles in several ways. First, changes in aerosol particle number, composition, and size distribution affected the size and lifetime of liquid, ice, and graupel hydrometeor particles (first indirect, or microphysical effect). Second, absorption by black carbon within and between cloud and precipitation hydrometeor particles heated clouds, causing evaporation and increasing stability below the cloud (radiative effect). Thus, the model was able to treat the “boomerang effect” [Koren *et al.*, 2008], whereby an increase in aerosol at low aerosol level increases cloud fraction and an increase in aerosol at high aerosol level decreases cloud fraction. Changes in aerosol also affected precipitation, which fed back to the removal rate of aerosol particles themselves (“Rainout effect”) [Jacobson, 2002b].

Carbon dioxide, like other gases, affected irradiances and radiative heating rates as it is an IR absorber. Its absorption affected the temperature at all altitudes and horizontal locations, and changes in temperature affected stability, local temperature and pressure gradients (thus wind speeds and the transport of all gases and particles), evaporation rates from oceans, lakes, soils, and clouds, and the temperature-dependence of natural emissions, air chemistry, and particle microphysics.

Changes in CO<sub>2</sub> also affected CO<sub>2</sub> photosynthesis (which depended on CO<sub>2</sub> backpressure) in chlorophyll and green plants, thus chlorophyll and leaf respiration rates. Changes in CO<sub>2</sub> further changed dissolution and evaporation rates of CO<sub>2</sub> into the ocean (which were also affected by the changes in temperature due to CO<sub>2</sub>), CO<sub>2</sub> weathering rates, ocean pH and chemical composition, sea spray pH and composition, and rainwater pH and composition. Changes in sea spray composition, in turn, affected the radiative properties of sea spray, thus radiative heating rates.

### **S3. Description of Simulations**

The model was run in one-way nested mode from the global to local scale on three sets of domains: one nested from the globe to California (California simulation), a second nested from the globe to California to the South Coast Air Basin (SCAB simulation), and a third nested from the globe to the United States (US simulation). The domain horizontal resolutions were as follows: global (4°-SN x 5°-WE resolution), United States (0.5° SN x 0.75° WE with the southwest corner grid cell centered at 22.0°N and -126.5° W and 58 SN cells x 84 WE cells), California (0.2° SN x 0.15° WE ≈ 21.5 km x 14.0 km with the southwest corner grid cell centered at 30.0°N and -126.0° W and 60 SN cells x 75 WE cells), and SCAB (0.045° SN x 0.05° WE ≈ 4.7 km x 5 km with the southwest corner grid cell centered at 30.88°N and -119.35° W and 46 SN cells x 70 WE cells)

The global domain included 47 sigma-pressure layers up to 0.22 hPa (≈60 km), with high resolution (15 layers) in the bottom 1 km. The nested regional domains included 35 layers exactly matching the global layers up to 65 hPa (≈18 km). The model was initialized with 1-degree global reanalysis data but run without data assimilation, nudging, or model spinup. The nesting time interval for passing meteorological and



chemical variables was one hour in all cases. All physical processes described, including emissions, were solved in all nested domains, including the global domain, of all simulations.

Four pairs of simulation were run: two for the California domain (starting in 2006 and 2007, respectively, to check interannual variability and the robustness of the result for the same domains across different initialization periods), one for the Los Angeles domain, and one for the U.S. domain. Baseline and sensitivity simulations were run for February-April 2006 and August-October 2006 in the global-through-Los Angeles domains and for January-March 2006 and July-September 2006 in the global-through-U.S. domains. For each sensitivity simulation, only the finest-resolved domain was perturbed (e.g., by removing CO<sub>2</sub> emissions) to minimize any impacts of perturbations to the coarse-resolution domains that might feed into the finest-resolved domains.

In some simulation results (e.g., California-domain results in Figure 2 of the main text), a checkerboard pattern can be seen in some parameters over the ocean. This arose because slight differences in wind speed (such as due to an enhanced or decreased sea breeze from localized land surface heating or cooling, respectively), changed the emission rate of wind-driven sea spray and its components (listed in the footnote of Table S1 as H<sub>2</sub>O, Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>, Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, H<sub>2</sub>SO<sub>4</sub>(aq), HSO<sub>4</sub><sup>-</sup>, and SO<sub>4</sub><sup>2-</sup>), as indicated by the fact that the PM<sub>2.5</sub> figure (2g) has the greatest checkerboard change. Subtle differences in the relative humidity in each cell then affected the water uptake by aerosol particles of different size and composition differentially in each cell, which affected gas uptake by the particles by dissolution and condensation. Acid gas uptake (e.g., sulfuric and nitric acid) near the coast in particular, then reduced the Cl<sup>-</sup> concentration through sea-spray acidification. The changes in the number and sizes of aerosol particles affected cloudiness, which fed back to surface solar radiation, temperatures, wind speeds, and water evaporation. Since cloudiness is more significant over the water than over land, since the relative humidity is higher over the ocean than over land, and since wind-driven sea spray particles are affected significantly by the relative humidity, subtle spatially-varying changes in wind speed and relative humidity over the ocean are expected to have spatially-varying impacts on aerosol and cloud properties there.

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