

Using synthetic tracers as a proxy for summertime PM_{2.5} air quality over the Northeastern United States in physical climate models

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[1] Fine particulate matter (PM_{2.5}) is a criteria pollutant. Its sensitivity to meteorology implies its distribution will likely change with climate shifts. Limited availability of global climate models with full chemistry complicates efforts to assess rigorously the uncertainties in the PM_{2.5} response to a warming climate. We evaluate the potential for PM_{2.5} distributions in a chemistry-climate model under current-day and warmer climate conditions over the Northeastern United States to be represented by a Synthetic Aerosol tracer (SA_t). The SA_t implemented into the Geophysical Fluid Dynamics Laboratory chemistry-climate model (AM3) follows the protocol of a recent multimodel community effort (HTAP), with CO emissions, 25-day chemical lifetime, and wet deposition rate of sulfate. Over the Northeastern United States, the summer daily time series of SA_t correlates strongly with that of PM_{2.5}, with similar cumulative density functions under both present and future climate conditions. With a linear regression model derived from PM_{2.5} and SA_t in the current-day simulation, we reconstruct both the current-day and future PM_{2.5} daily time series from the simulated SA_t. This reconstruction captures the summer mean PM_{2.5}, the incidence of days above the 24-h mean PM_{2.5} NAAQS, and PM_{2.5} responses to climate change. This reconstruction also works over other polluted Northern Hemispheric regions and in spring. Our proof-of-concept study demonstrates that simple tracers can be developed to mimic PM_{2.5}, including its response to climate change, as an easy-to-implement and low-cost addition to physical climate models that should help air quality managers to reap the benefits of climate models that have no chemistry.

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1. Introduction

[2] Fine particulate matter (PM_{2.5}) causes a variety of adverse health effects, resulting in increased mortality and hospital admissions for cardiovascular and respiratory diseases [Dockery *et al.*, 1993; Pope, 2000; Pope *et al.*, 2002; Schwartz *et al.*, 2002; Pope and Dockery, 2006; Parry *et al.*, 2007]. It is also responsible for a reduction in visibility [Park *et al.*, 2006] and changes in climate [Parry *et al.*, 2007; Kloster *et al.*, 2010]. In the United States, PM_{2.5} is regulated under the Environmental Protection Agency's National Ambient Air Quality Standard (NAAQS) as a criteria air pollutant with a daily mean threshold of 35 $\mu\text{g}/\text{m}^3$ and an annual mean threshold of 15 $\mu\text{g}/\text{m}^3$. As the concentration of PM_{2.5} is highly sensitive to meteorological conditions, it is expected to respond to climate change [Jacob and Winner, 2009; Fiore *et al.*, 2012]. Therefore, regardless of potential changes in precursor emissions, a thorough understanding of the effects of global climate change on PM_{2.5} concentrations is central to future air quality planning. We show that implementing synthetic tracers into the current generation of climate models offers a powerful, simple, and computationally efficient approach to project climate-driven changes to air quality.

[3] Although previous studies project significant changes in PM_{2.5} in a warming climate, uncertainties exist regarding even the sign of the change ($-0.1 \pm 10 \mu\text{g}/\text{m}^3$) [Jacob and Winner, 2009]. Recent studies using coupled chemistry-climate models suggest an increase in PM_{2.5} despite globally increasing precipitation, reflecting a reduction in large-scale precipitation over polluted regions and seasons [Fang *et al.*, 2011; Kloster *et al.*, 2010]. On the regional scale, changes of PM_{2.5} in a warming climate are even more difficult to project due to uncertainties in regional boundary layer ventilation and precipitation [Meehl *et al.*, 2007]. As only a handful of climate models are available with fully coupled chemistry and computational constraints often limit chemical transport models to using a limited set of meteorology from a single climate model, a rigorous evaluation of the uncertainties in the PM_{2.5} response to various climate change scenarios projected by a suite of climate models is challenging.

[4] Synthetic tracers have been applied previously within chemical transport models to advance our understanding of the source, transport, and removal processes for regional air pollution and hemispheric pollution transport [e.g., Bey *et al.*, 2001; Staudt *et al.*, 2001; Stohl *et al.*, 2002; Liu *et al.*, 2005; Shindell *et al.*, 2008; HTAP, 2007, 2010]. Synthetic tracers have also been used in climate models to estimate how changes in the atmospheric circulation and precipitation affect air quality [Mickley *et al.*, 2004; Fang *et al.*, 2011]. Over many regions, including the Northeastern United States, the day-to-day variability of PM_{2.5}, which is directly

related to pollution episodes, is strongly affected by synoptic transport processes [Cooper *et al.*, 2001; Tai *et al.*, 2012; Appelhans *et al.*, 2012], and wet removal dominates PM_{2.5} loss [Textor *et al.*, 2006]. Therefore, it is important to include both transport and wet removal in the design of a synthetic tracer for PM_{2.5}. The Task Force on Hemispheric Transport of Atmospheric Pollution (HTAP) has designed a diagnostic tracer experiment (TP1x), consisting of a soluble tracer with CO emissions to compare aerosol simulations across models. We incorporate a similar diagnostic tracer into the Geophysical Fluid Dynamics Laboratory (GFDL) AM3 model and provide a proof-of-concept study that illustrates the potential of synthetic tracers to represent daily PM_{2.5} statistics, including their changes in a warming climate.

2. Method

2.1. Model and Simulations

[5] We use simulations from the GFDL chemistry-climate model (AM3) for current-day (1981–2000) and future (2081–2100) climate conditions and PM_{2.5} distributions as described by Fang *et al.* [2011]. Briefly, the simulations are designed to study the effect of climate change on pollutants in the absence of emission changes. We further aim to minimize “noise” from interannual variability in order to allow the “signal” from climate change to emerge in our simulations. Specifically, the current-day simulation is driven by a 1981–2000 climatology of observed monthly mean sea surface temperature and sea ice. The future simulation is driven by this observed climatology plus the 19-model (from IPCC AR4) ensemble mean difference of 20-year average values (applied separately for each month, ensemble model mean annual change in SST is shown in Figure S1 in the Supporting Information) from 1981–2000 to 2081–2100 under the SRES A1B scenario. The global mean surface temperature increase projected by the IPCC AR4 model ensemble is 2.8 K. Emissions of short-lived pollutants in both present and future simulations are set to 1990 levels, repeated annually. The concentrations of the long-lived greenhouse gases (CO₂, N₂O, and CFCs) are set to 1990 values for the present but 2090 (A1B) values for the future simulation. CH₄ is set to the 1990 level for tropospheric chemistry calculations in both simulations but to the 1990 and A1B 2090 level for radiation (climate forcing) in the current-day and future simulations, respectively. Each simulation is run for 20 years following a 1-year spin-up.

[6] As CO is considered a useful tracer of anthropogenic pollutants, HTAP [2010] proposed synthetic tracers with 25-day chemical lifetime that follow CO emissions (including anthropogenic and biomass burning emissions from the RETRO [Schultz and Rast, 2007] and GFED v2 [van der Werf *et al.*, 2006] inventories for 2001, respectively) to help diagnose the transport processes affecting air pollutant distributions. We use these emissions and follow the HTAP approach of implementing a soluble version of this tracer, with wet deposition rates as sulfate, for our Synthetic Aerosol tracer (SA_t). The global lifetime of SA_t is about 4–6 days depending on season, close to that of the major PM_{2.5} components: the AEROCOM all-model-average annual lifetime is 4 days for SO₄ and dust and 6–7 days for primary organic matter and black carbon [Textor *et al.*, 2006]. In this study, we incorporate SA_t into the AM3 simulations and examine its utility as a proxy for PM_{2.5} air quality in physical climate models,

focusing mainly on the Northeastern United States. SA_t does not have real chemistry; therefore, it is expected to work best for PM_{2.5} components with little chemistry. Nevertheless, as we show in section 3, the capability of SA_t to represent the daily variability of and climate change effects on PM_{2.5} suggests that transport and wet deposition are the major drivers for the climate change effect and the daily variability.

2.2. Model Evaluation

[7] Donner *et al.* [2011] show that the AM3 simulated aerosol optical depth and co-albedo broadly capture those in AERONET observations (agreement within a factor of 2 at most sites, with a global correlation coefficient ranging from 0.6 to 0.7). As we focus here on air quality over the Northeastern United States in summer (June–July–August), we evaluate the surface PM_{2.5} concentration (defined as the sum of sulfate, nitrate, dust with a dry radius less than 0.1 μm, secondary organic carbon, sea salt with a dry radius less than 0.2 μm, black carbon, and organic carbon; dry radius thresholds are chosen to ensure the aerosol size is less than 2.5 μm when water content is included) in the current-day simulation with the U.S. Air Quality System (USAQS) PM_{2.5} observations.

[8] Our current-day simulation is designed to represent the climatological mean situation during 1981–2000. We thus evaluate the simulated summer mean and seasonal cycle of PM_{2.5} with observations from the U.S. Air Quality System (AQS) averaged from 1997 (the earliest year with PM_{2.5} data reported) through 2007. The simulated summer mean PM_{2.5} is spatially correlated with the AQS observations ($R \sim 0.7$, over the Northeastern United States) with an average bias of 34% (Supporting Information, Figure S2). The positive bias occurs mainly over the Northeastern United States, where sulfate aerosol contributes above 50% to PM_{2.5} in the model. This bias is expected since the simulation applies emissions for the year 1990 throughout the 20-year simulation (to minimize climate noise; section 2.1) but U.S. emissions of sulfur dioxide (SO₂, precursor to sulfate aerosol) decreased by about 30% from 1990 to 2000 [EPA, 2001], and this decrease should be reflected in the 1997–2007 AQS observations. The model exaggerates the seasonal cycle of simulated PM_{2.5} over the Northeastern United States, likely due to excessive SO₂ emissions and subsequent sulfate production, but captures the summertime maximum (Supporting Information, Figure S3) which reflects stronger chemical production of aerosols during this season.

3. Representing the PM_{2.5} Distribution with a Synthetic Tracer (SA_t)

[9] The daily time series of SA_t correlates strongly with PM_{2.5} during summer ($R=0.86$ for all 20 summers and $R=0.8–0.9$ during individual summers). The cumulative density functions (CDFs) of daily mean surface concentrations of SA_t and PM_{2.5} for current-day and future climate conditions over the Northeastern United States (NEUS) suggest that both PM_{2.5} and SA_t have higher values at the high and low tails than a normal distribution (Figures 1a and 1b). Over the 10–98 percentile range, the SA_t and PM_{2.5} distributions are similar (Figure 1c). In the warmer climate, the mean summer PM_{2.5} concentration increases by 15% (from 20 μg/m³) from the current day to the future simulation, while the SA_t concentration increases similarly, by 16%. Both distributions indicate an increase, especially at the upper tail of the distribution.

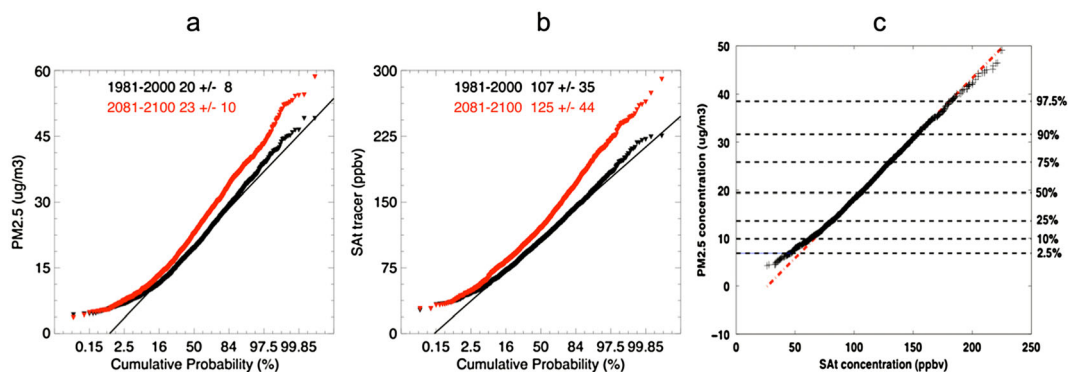


Figure 1. Cumulative density functions of summertime (a) daily PM_{2.5} and (b) a SAT tracer with 25-day chemical lifetime and wet deposition in the GFDL AM3 current-day (black) and future (red) simulations. Each distribution is constructed from 92 summer daily average values over the U.S. Northeast region (36–46 N, 70–80 W, land only, 15 grid cells total, see Figure S2 in the Supporting Information) times 20 years each for 1990 and 2090 climate conditions. The numbers in the panels correspond to the mean and standard deviation of the distributions; the black solid line indicates a normal distribution with the same mean and standard deviation of PM_{2.5} (in Figure 1a) and SAT tracer (in Figure 1b). (c) Quantile-quantile plot of summer daily SAT and PM_{2.5} concentrations. Red line is the reference showing the same distribution. Dashed black lines indicate PM_{2.5} at 2.5, 10, 25, 50, 75, 90, and 97.5% quantiles.

Concentrations at the upper tail (above 75%, representing pollution episodes) increase by 19% and 15% in the future climate for SAT and PM_{2.5}, respectively, while the increases at the lower tail (below 10%) only increase by 12% and 10%. The change from the current day to the future is statistically significant ($p < 0.01$, assessed by Kolmogorov-Smirnov test) for both SAT and PM_{2.5}. The stronger increase in the upper tail, which occurs for both SAT and PM_{2.5}, is consistent with the findings of *Mickley et al.* [2004] that climate change may increase high pollution episodes more strongly than mean pollution levels. As SAT is only affected by changing transport and precipitation in a changing climate, this higher tail increase is driven by changes in those two factors. Decreasing cyclones (hence increased stagnation, consistent with *Mickley et al.* [2004]) and decreasing frequencies of precipitations (especially of low-intensity precipitation, as in *Fang et al.* [2011]) are the major drivers for the higher tail increase. The strong correlation of daily average SAT and PM_{2.5} and similarity of their CDFs suggest that the day-to-day variations of PM_{2.5} are mainly driven by transport and removal and that SAT may be used as an index to reconstruct PM_{2.5} and its changes in a warmer climate.

3.1. Linear Regression Model to Reconstruct PM_{2.5} from SAT

[10] The strong correlation between PM_{2.5} and SAT motivates us to examine the extent to which we can use a linear regression model to reconstruct daily PM_{2.5} concentrations from daily SAT concentrations, i.e., $C_{\text{PM}_{2.5}} = a + b * C_{\text{SAT}}$. Applying daily current-day summer concentrations of SAT and PM_{2.5} in the AM3 simulations, we derive the coefficients: $a = -1.6$ ($-0.8, -2.5$) $\mu\text{g}/\text{m}^3$ and $b = 0.20$ ($0.20, 0.21$) $\mu\text{g}/\text{m}^3$ per ppbv (with numbers in the brackets indicating the 99% confidence interval of these coefficients). The linear model captures the magnitude of the 20-summer mean PM_{2.5} concentrations ($19.8 \mu\text{g}/\text{m}^3$ for the reconstruction versus $20.2 \mu\text{g}/\text{m}^3$ for simulated PM_{2.5}) and also 74% of the total PM_{2.5} daily variability under current climate conditions (Figure 2a). Comparison of daily time series further confirms the utility of this linear regression model in representing the daily time series of PM_{2.5} concentration over the Northeastern United States

(Figure 2b). During days with PM_{2.5} episodes, the reconstruction tends to underestimate simulated PM_{2.5} (section 3.2).

[11] We apply the same method to SAT and PM_{2.5} in the future simulation and derive similar coefficient values, i.e., $a = -1.2$ ($-0.6, 2.2$) $\mu\text{g}/\text{m}^3$ and $b = 0.20$ ($0.19, 0.20$) $\mu\text{g}/\text{m}^3$ per ppbv (with numbers in the brackets indicating the 99% confidence intervals of these coefficients), which confirms that this relationship between SAT and PM_{2.5} derived from the current-day simulation is also robust under the future climate scenario. This equivalence implies that the SAT relationship derived from the current-day simulation offers a good representation of PM_{2.5} even under climate change scenarios.

[12] We next use the coefficients of the current-day linear regression model along with future SAT, all that would be available from implementing the synthetic tracer in a physical climate model, to reconstruct future PM_{2.5}. The reconstructed values are biased less than 2% for the 20-summer mean PM_{2.5} concentration and capture 80% of the AM3 simulated PM_{2.5} daily variability in the future simulation (Figures 2c and 2d). The future change in mean PM_{2.5} is also fairly well captured by applying the linear regression model ($3.5 \mu\text{g}/\text{m}^3$ increase in the linear regression model versus $3 \mu\text{g}/\text{m}^3$, simulated by AM3). The PM_{2.5} non-attainment days corresponding to the $35 \mu\text{g}/\text{m}^3$ daily mean PM_{2.5} standard are also well represented: 4% and 14% of all summer days are non-attainment days in AM3 present and future climate simulations, respectively; those statistics are 3% and 12% for the regression model.

3.2. Factors Leading to Underestimated SAT-derived PM_{2.5} Values on Polluted Days

[13] Despite the high correlation between the SAT and PM_{2.5} time series, Figures 2b and 2d show that the linear regression model slightly underestimates PM_{2.5} concentrations relative to the AM3 simulation. We further examine these situations in the current-day simulation by comparing mean PM_{2.5} and precipitation distributions on all summer days and on days when the bias between the simulated and reconstructed PM_{2.5} exceeds $10 \mu\text{g}/\text{m}^3$. The largest underestimates typically occur when PM_{2.5} over the Northeastern

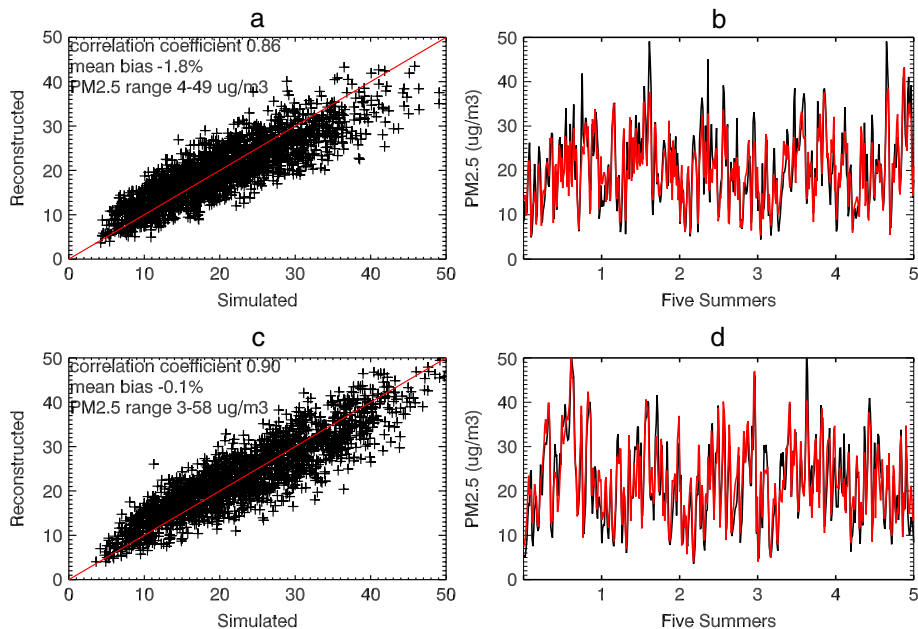


Figure 2. (a) Scatter plot of daily PM_{2.5} as simulated by the GFDL AM3 model versus PM_{2.5} as reconstructed by applying the linear regression model (section 3.1) to the SAT distribution over the Northeastern United States for the 20 summers with 1990 climate conditions (black plus signs). The red line shows the 1:1 relationship. (b) An example 5 year time series of simulated (black) and reconstructed (red) daily PM_{2.5} concentrations over the Northeastern United States (unit: $\mu\text{g}/\text{m}^3$); (c and d) similar to Figures 2a and 2b, respectively, but with 2090 climate condition.

United States is larger than the summer average (by $\sim 10 \mu\text{g}/\text{m}^3$; Supporting Information, Figures S4a and S4b) and when precipitation is stronger than average (Figures S4c and S4d). Results are similar for composite plots from the future simulations (not shown).

[14] Further analysis indicates that these precipitation events are associated with pollution transport. For example, for one such episode, PM_{2.5} transported from Kentucky and Tennessee enhances the PM_{2.5} concentration over the Northeastern United States. Inspection of daily SAT distributions over the Eastern United States during this event reveals negligible contribution of transport from these regions to the SAT concentration over the Northeastern United States (Supporting Information, Figure S5) for two reasons. First, the SAT tracer emission follows CO; its source over Kentucky and Tennessee is much weaker compared to that of PM_{2.5} precursors (SO₂, for example, Supporting Information, Figure S6). Second, the SAT tracer is soluble, while some components of PM_{2.5} and its precursors, such as hydrophobic black carbon (BC) and organic carbon (OC), are not soluble. Consequently, on days with strong precipitation, SAT is removed more quickly than PM_{2.5}. These findings imply that the utility of SAT may be further improved by implementing two separate tracers: one with SO₂ emissions and sulfate solubility (as tested in section 4) and one with carbonaceous emissions and solubility.

3.3. Extension to Springtime and Other Regions

[15] To examine whether SAT can represent PM_{2.5} over other polluted regions at northern midlatitudes, we analyze SAT and PM_{2.5} daily time series for 20 summers over the Midwest United States (MWUS), Northern East Asia (NEEA), Southern East Asia (SEEA), Mediterranean Europe (MEU), and Northern Europe (NEU). In general, SAT is strongly correlated with PM_{2.5} ($R=0.71\sim 0.89$), although

the regression slope varies by region (slope= $0.19\sim 0.31$), reflecting differences in regional budgets of PM_{2.5} (Supporting Information, Table S1). As long as region-specific relationships (regression coefficients) are determined from a current-day simulation, future PM_{2.5} can be reconstructed fairly well by using the current-day linear relationship between SAT and PM_{2.5} along with the simulated future SAT tracer (Table S1). Over all regions, the mean bias in PM_{2.5} is within $\pm 7\%$ and about 50–80% variability of daily PM_{2.5} is captured.

[16] We further analyze the utility of SAT in spring (MAM) when long-range transport is relatively more important than during summer (e.g., HTAP [2010]). Over NEUS, MWUS, MEU, and NEU, simulated PM_{2.5} and SAT are correlated in the current-day simulations slightly less than in summer ($R=0.68\sim 0.75$, Table S1). The reconstructed and simulated future PM_{2.5} are also fairly well correlated ($R^2\sim 0.5$), and the mean bias of this regression model is within $\pm 10\%$ over these regions (Table S1). Over NEEA and SEEA, however, the springtime correlation is weak ($R^2 < 0.2$), indicating that the SAT tracer does not adequately represent PM_{2.5}. The poor applicability of SAT over SEEA and NEEA is associated with stronger long-range transport during spring. As shown in section 3.2, discrepancies in solubility and emissions between SAT and PM_{2.5} precursors may accumulate during long-range transport, weakening the correlation between them. This problem is largely overcome by tying SAT to SO₂ rather than CO emissions (section 4). We conclude that synthetic tracers can be generally applied in climate models with similar success in representing PM_{2.5} over several Northern Hemispheric regions in spring and summer.

4. Sensitivity of Tracer Representation to Different Current-day Meteorology and Emissions

[17] Prior to implementing synthetic tracers for air quality applications in physical climate models, we need to

determine the extent to which the relationship between PM_{2.5} and SAT depends on model configuration. As a preliminary step, we analyze here a different version of the GFDL AM3 model in which the meteorology is nudged to National Centers for Environmental Prediction (NCEP) re-analysis winds as described by Lin *et al.* [2012a, 2012b] for the time period of 1979–2010 and thus is comparable to a chemical transport model. This simulation includes the same SAT tracer and applies annually invariant emissions of air pollutants during 1979–2010. Despite the different setting of AM3 nudged simulations, its SAT and PM_{2.5} are still strongly correlated with similar correlation coefficients ($R=0.71\sim 0.85$, Supporting Information, Table S2). The slopes, which are applied to the reconstruction, however, slightly vary due to differences in the emission inventory used (the nudged version uses climatological mean emissions from 1979 to 2010 [Lin *et al.*, 2012b], while the interactive version uses emissions representing 1990 levels [Fang *et al.*, 2011]).

[18] We further use this simulation to explore whether we can improve the SAT performance over regions where sulfate dominates total PM_{2.5} by tying the emissions to SO₂. For this purpose, we implement a second synthetic tracer, denoted SO₂t, which is the same as SAT except the CO emissions are replaced by SO₂ emissions. Given the larger molar emission rate of CO versus SO₂ (global emissions of SO₂ and CO are approximately 2 versus 44 Tmol per year, respectively) and hence more abundant SAT than SO₂t in the atmosphere, the slope of the regression model of SO₂t is larger than that for SAT in summer (slope = 1.54 ~ 2.25, Table S2). A strong linear correlation emerges for both the SO₂t and SAT tracers and PM_{2.5} over all regions ($R=0.71\sim 0.91$, Table S2). The linear correlation between SO₂t and PM_{2.5} is stronger than that between SAT and PM_{2.5} over all regions chosen, consistent with our hypothesis in section 3.2. During spring, the improvement is particularly large over NEEA and SEEA, where the correlation coefficients between tracers and PM_{2.5} increase from 0.39 and 0.40 (using SAT and PM_{2.5}) to 0.73 and 0.72 (using SO₂t and PM_{2.5}).

5. Conclusions

[19] We implement a simple soluble synthetic aerosol tracer (SAT; tied to CO emissions) in the GFDL AM3 coupled chemistry-climate model [Donner *et al.*, 2011] and show that it correlates strongly ($R=0.86$) with day-to-day variations in simulated surface PM_{2.5} concentrations over the Northeastern United States. With a simple linear regression model that uses SAT to reconstruct PM_{2.5}, we show that this tracer can represent the response of the regional PM_{2.5} distribution to climate change. Specifically, the PM_{2.5} distribution reconstructed from SAT captures around 80% of the daily variability, the magnitude of regional mean PM_{2.5}, and the incidence PM_{2.5} non-attainment days, and their changes from the current-day distribution over several Northern Hemispheric regions in both spring and summer (Table S1; Figures 1 and 2). The tracer is similarly accurate when applied in AM3 with a different configuration (meteorology and emissions; Table S2). Our proof-of-concept study demonstrates that, as long as the model-specific regression coefficients are determined from a full-chemistry simulation for a current-day scenario, future PM_{2.5} can be reconstructed

by applying the current-day linear relationship between synthetic tracers and PM_{2.5} to the simulated future synthetic tracer at least on regional scales (as seen from AM3 in Table S1). Further refinements may improve the utility of synthetic tracers over specific regions of interest (e.g., tracers designed for sulfate, carbonaceous aerosols, or dust).

[20] Future chemistry-climate multimodel ensemble projects could include synthetic tracers (as in HTAP [2010]) to evaluate whether model-independent relationships between tracers and PM_{2.5} can be developed. Such an effort is an essential step towards eliminating the need for model-specific regression coefficients to be determined from a current-day full-chemistry simulation. Our method would then be an easy-to-implement and low-cost addition to state-of-the-art physical climate models, such as those routinely run in centuries long “production” simulations for assessment reports at international modeling centers. Trade-offs between increasing spatial resolution, important for representing smaller scale processes and additional process-level complexity, such as full representation of atmospheric chemistry, are inevitable. Implementing such a tracer into high-resolution physical climate models would expand the number of scenarios and simulation years available to assess multimodel air quality projections, allowing air quality managers to reap the benefits of these models—which would otherwise not be possible in the absence of fully coupled chemistry.

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