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Dr. Naresh Kumar
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Dear Naresh:

I write to propose a continuation of my current 2-year project with EPRI, “**Natural Visibility Conditions in the United States and the Effect of Transboundary Pollution**” (current funding period 2/15/02-2/14/04). The goals of this project are to (1) better quantify natural PM concentrations in the U.S. for application of the EPA Regional Haze Rule (RHR), and (2) quantify the role of transboundary pollution in increasing baseline PM concentrations above natural values. We define here *baseline* concentrations as those not amenable to domestic emission controls, including contributions from both natural sources and transboundary pollution. Our analysis is based on a global 3-dimensional model of atmospheric composition (GEOS-CHEM) driven by NASA/GEOS assimilated meteorological observations and applied to simulations of sulfate, nitrate, OC, EC, and dust. The model is evaluated with observations from the IMPROVE and other networks to test our understanding of sources and other processes determining PM concentrations. Sensitivity simulations are used to quantify the contributions from natural and transboundary pollution sources to the PM loadings. Our simulations so far have used the GEOS-CHEM model with 2°x2.5° horizontal resolution. We propose in future work to use a nested version of the model with 1°x1° resolution over North America. In this letter I briefly summarize our accomplishments so far and outline a plan of work for the next two years. A total of \$213,000 is requested for the two years (2/15/04-2/15/06).

Summary of Accomplishments. Our work so far has focused on (1) general evaluation of the model PM simulation using IMPROVE and other data, (2) application of the model to quantify mean natural and baseline PM concentrations in the eastern and western U.S. for comparison to Table 2.1 of the EPA [2001] RHR document. We have

investigated carbonaceous (OC/EC), sulfate-nitrate-ammonium, and dust PM components separately.

Our OC/EC work was published recently in *J. Geophys. Res.* [Park et al., 2003]. We started from our best *a priori* understanding of OC and EC sources, including satellite data to constrain fire emissions, and found that the model had substantial skill in reproducing the observed geographical distribution of OC and EC at IMPROVE sites for 1998 ($R^2 = 0.67$ for OC, 0.84 for EC), with small but significant biases (-26% for OC, -15% for EC). We optimized the individual sources of OC and EC within their accepted ranges of uncertainties to fit the IMPROVE data and remove the biases. Our resulting baseline OC and EC concentrations are 2-3 times higher than the default EPA natural values, except for OC in the east for which there is good agreement. Most of the discrepancy is due to fire emissions, including major contributions from Canada and Mexico. Transboundary transport of pollution is inconsequential.

Our sulfate-nitrate-ammonium work is presently being prepared for publication; it was reported at the August RPO telecon and at the Gordon Research Conference. Model results were evaluated with 1998 and 2001 observations from networks in the U.S. and Europe (IMPROVE, CASTNET, NADP, EMEP). Sulfate results show high correlations with observations (R^2 typically 0.9) and no significant bias except in summer (30% negative bias). Natural levels of sulfate and nitrate in the model are consistent with the EPA default estimate, but transboundary transport of pollution from Canada, Mexico, and Asia elevates baseline concentrations by a factor of 3 above these natural levels. Transpacific transport of Asian anthropogenic aerosol accounts for 30% of baseline sulfate in both the western and eastern U.S. Using our baseline concentrations to define the 2064 endpoint of the RHR, rather than the EPA natural defaults, would greatly decrease the level of emission controls needed in phase I implementation (2004-2018) of the RHR.

Our dust work has initially applied the dust source scheme of Ginoux et al. [2001] to a simulation of the IMPROVE data for 2001. The model demonstrates skill in reproducing the transpacific transport of Asian dust, including the major episode of mid-April 2001. However, background concentrations at western sites are overestimated, implying an excessive domestic source. We are presently implementing an improved dust mobilization scheme [Zender et al., 2001] that we expect will correct the problem.

Our agenda from now to the end of the project (2/14/04) is to (1) complete the sulfate-nitrate paper, (2) correct the dust simulation, and (3) prepare the final report. We will also start work on improving the simulations of OC and sulfate-nitrate to address identified flaws, but we expect this work to be completed as part of the proposed project and discuss it further below..

Proposed work. Our work so far has provided an assessment of mean baseline PM concentrations in the U.S. including the contributions from natural and transboundary pollution sources. The implications for the RHR are potentially important. Evaluation of model results with observations has shown substantial success for seasonal mean aerosol concentrations and spatial distributions. However, there are flaws that need to be corrected. Also, for application to the RHR we need to examine more pertinent model diagnostics. These include frequency distributions of baseline and natural visibility at the IMPROVE

sites, since the RHR is based on statistics for the 20% worst visibility days. We also need to provide more geographical specificity to our results, going beyond the simple “east” and “west” classification.

We must increase confidence in our estimates and provide uncertainty ranges. Model evaluation with seasonal mean concentrations provides only a limited test of our ability to simulate baseline PM concentrations. We propose to use the frequency distribution of concentrations at IMPROVE sites, as well as the correlations between species, as a test of the model. We also propose to use the MODIS satellite observations of aerosol optical thickness (AOT) as an important constraint for our ability to reproduce PM concentrations over the range of conditions found across the United States including transboundary and transpacific transport.

We further propose to examine the effects of global changes in emissions and climate on future baseline PM concentrations. These effects are important to address from a 2064 endpoint perspective. We will take advantage here of a new EPA-funded project, Global Change Impacts on Air Quality (GCIAQ), that is aimed at assessing the impact of 2000-2050 changes in climate and global emissions on the achievability of ozone and PM primary air quality standards. This project was initiated in 2003 for a 3-year period and involves collaboration of groups at Harvard (D.J. Jacob, P.I.), Caltech (J.H. Seinfeld), NASA/GISS (D. Rind), Argonne National Lab (D.R. Streets), U. Tennessee (J. Fu), EPA/ORD (A. Gilliland) and EPA/OAQPS (C. Jang). It will use climate projections from the GISS general circulation model and future emission forecasts (including anthropogenic sources and fires) from D.R. Streets. Two different climate forcing and emission scenarios will be considered, one optimistic and one pessimistic, drawing on the A1 and B1 scenarios of IPCC [2001]. The GISS GCM meteorological fields and the future emission estimates will be implemented in a GEOS-CHEM coupled PM-oxidant simulation to diagnose global changes in atmospheric composition and implications for regional air quality. We will use this simulation to investigate trends in baseline PM concentrations from 2000 to 2050. We recognize that the endpoint for the RHR is 2064, not 2050, but there is little confidence to be had in global change scenarios beyond 2050 and substantial effort would be involved in extending the GCIAQ simulations to 2064.

We propose therefore to focus our continuation of this project on four objectives:

- (1) Correct known model flaws and increase model resolution;
- (2) Provide our results in spatial and statistical forms that are specifically pertinent to the RHR;
- (3) improve confidence in our baseline PM estimates, and define uncertainty ranges, through more extensive evaluation with IMPROVE observational statistics and MODIS satellite observations;
- (4) assess the effects of future global changes in climate and emissions on baseline PM concentrations in the U.S.

We describe below the specific tasks for addressing each objective. Tasks 1-3 will use a nested, version of the GEOS-CHEM model with 1°x1° horizontal resolution over North America and 4°x5° for the rest of the world. The GEOS meteorological data are originally available with 1°x1° resolution; the 1-way nested capability from 4°x5° to 1°x1°

resolution is already in place in GEOS-CHEM and is presently being applied to coupled oxidant-PM simulations for North America in support of the summer 2004 ICARTT aircraft campaigns. Execution of tasks 2-3 will involve a 5-year GEOS-CHEM model simulation for 1998-2002 to (1) match the RHR requirement for characterizing the frequency distributions of visibility at IMPROVE sites, (2) improve concentration statistics for model evaluation, particularly for the interannually varying fire and dust sources.

Task 1. Correct known model flaws and increase model resolution. We will correct flaws in emissions and processes revealed in our previous model evaluation:

- (1) *Overestimate of OC in the northwest.* This reflects an overestimate of the regional biogenic source [Park et al., 2003]. Our current formulation of this source assumes a uniform 10% yield from the oxidation of terpenes. However, this yield is known to decrease with decreasing total aerosol mass concentration, reflecting the partitioning of the terpene oxidation products between the gas and aerosol phases [Griffin et al., 1999]. Lower aerosol concentrations in the northwest imply lower OC yields from terpene oxidation. We will implement such a dependence in the model. As part of the GCIAQ project, John Seinfeld's group at Caltech will be implementing into GEOS-CHEM a detailed mechanistic simulation of secondary OC [Chung and Seinfeld, 2002] and we will adopt it as it becomes available.
- (2) *Overestimate of ammonium in fall.* This flaw likely reflects a seasonal overestimate of ammonia emissions in current inventories, as demonstrated by Gilliland et al. [2003a] in inverse modeling of the NADP NH_4^+ wet deposition data. Incorporation into the CMAQ regional model of the seasonal cycle derived from this inverse analysis has been found to improve greatly the simulations of both ammonium and nitrate aerosol [Gilliland et al., 2003b]. We will adopt it in our GEOS-CHEM simulation.
- (3) *Underestimate of sulfate in summer.* Excessive scavenging of SO_2 in wet convective updrafts appears to be a major cause. In the present version we scavenge SO_2 quantitatively in convective updrafts, limited only by the supply of H_2O_2 entrained in the updraft. However, kinetic limitations to the aqueous-phase oxidation of SO_2 by H_2O_2 , as well as scavenging of H_2O_2 , can greatly reduce the SO_2 scavenging efficiency [Mari et al., 2000]. We will improve our convective scavenging module to account for these effects.

Other recognized flaws in the model appear to reflect spatial resolution, including coastal and topographical effects, and the definition of fire sources and plumes [Park et al., 2003]. Increasing the resolution to $1^\circ \times 1^\circ$ should greatly improve these aspects of the simulation. We will repeat our 1-year simulation for 2001 at that resolution and evaluate the model with observed seasonal mean PM concentrations, as done before, to verify the expected improvements.

Task 2. Provide model results in form most pertinent to the RHR

Application of the RHR uses as metric the 20% worst visibility days (i.e., the 20th percentile of the visibility frequency distribution) at the individual IMPROVE sites. The EPA [2001] document recommends that the 20th percentile of the natural visibility frequency distribution be calculated by taking the mean natural visibility (deciview units) derived from the mean default natural PM concentrations, and assuming a lognormal distribution for this natural visibility with geometric standard deviations of 2 in the west and 3 in the east, based on the work of Aves and Malm [2000] as cited by EPA[2001]. We will compare this approach to the frequency distribution statistics for baseline and natural visibility obtained from our model simulation. Disturbances from fires, dust events, and transboundary pollution transport may lead to departures from the lognormal distribution. In addition, we expect the standard deviations to be more variable than the simple separation between ‘east’ and ‘west’ used in the EPA [2001] document.

We will also go beyond the simple classification of ‘east’ and ‘west’ in quantifying baseline PM concentrations at IMPROVE sites. Large differences between sites may be expected in terms of the different natural and transboundary pollution sources, and these need to be resolved if the implementation of the RHR is to be improved. We will extract from our model the baseline and natural PM concentrations at individual IMPROVE sites, and provide an analysis of the factors driving their spatial variability.

Task 3. Improve confidence in our baseline PM estimates and provide uncertainty ranges. We will use the frequency distributions of the concentrations of individual PM components at the IMPROVE sites to improve our testing of the sources contributing to baseline and natural concentrations in the model. Seasonal mean concentrations, which have been our focus so far, are often dominated by anthropogenic source contributions, and hence provide only a limited test of natural and transboundary sources. The low tail of the frequency distribution can provide better constraints in that regard, although care has to be taken to account for precipitation scavenging as a confounding factor. This can be done by using precipitation data from nearby NWS sites to screen specific days.

Interannual variability of OC and EC concentrations will provide an important test of our ability to describe fire emissions. Our specification of fire emissions uses a $1^{\circ} \times 1^{\circ}$ inventory of burned carbon developed in our group by Jennifer Logan and colleagues (Duncan et al. [2002]). This inventory combines information on land types and typical burning frequency with monthly satellite fire data. It was designed for global application and has been shown to provide a good simulation of the interannual variability of CO concentrations at remote NOAA/CMDL sites. Application to simulation of the IMPROVE observations for 1998 was also successful, as described by Park et al. [2003], capturing in particular the occurrences of large fires in Mexico in spring and in the western U.S. in summer. Evaluation against the interannual variability observed over a 5-year period will provide a more robust test and greatly increase confidence in our ability to quantify fire emissions in a manner useful to the application of the RHR. Any major difficulties in

simulating the IMPROVE observations will be discussed with Jennifer Logan in terms of possible improvements to the fire emission inventory.

Finally, we will evaluate the model with space-based observations of AOD from the MODIS instrument (<http://modis-atmos.gsfc.nasa.gov>) to provide a comprehensive view of PM concentrations in the U.S. including in the role of transboundary pollution. MODIS measures AOD by solar backscatter with a 10x10 km² field of view and achieves global coverage in 1-2 days. It has been launched on the Terra and Aqua satellites, which are in polar sun-synchronous orbits with daytime crossing times of 10:30 local and 1:30 local respectively. Validation with ground-based AOD measurements from the AERONET network indicates that MODIS has a sensitivity of 0.05 over land and 0.03 over ocean, which is good enough for the characterization of baseline PM concentrations and allows in particular the observation of transboundary and transpacific transport of pollution. We are already conducting general evaluations of GEOS-CHEM with MODIS over the eastern U.S. in preparation for the ICARTT aircraft campaigns in summer 2004. The proposed work will leverage from this activity.

The extensive evaluation of model results described here will allow us to place uncertainty ranges on our estimates of baseline and natural PM concentrations. We will do so by assessing how perturbations to individual sources affect PM concentrations in the model, and use the comparisons to observations as constraints to exclude source magnitudes outside a certain range. This is relatively straightforward since the relationships of PM components to their sources are roughly linear, with the notable exception of nitrate. For nitrate (which is only a minor contributor to baseline PM), we will make our best possible estimates of uncertainties on the basis of the ensemble of sensitivity simulations conducted.

Task 4. Assess the impacts of global changes in climate and emissions on baseline PM concentrations and visibility in the U.S.

We will compare results from our GCIAQ model simulations for 2000 and 2050 to examine the changes in baseline PM concentrations in the U.S. We will examine separately and in concert the effects of climate change and the effects of changes in emissions (including anthropogenic sources, fires, and biogenic processes). All model development and standard simulations will be leveraged from the GCIAQ project and will be at no cost to this project. We will need for this project to conduct specific sensitivity simulations, shutting off anthropogenic emissions globally and in the U.S. and North America, to identify natural and transboundary pollution contributions to PM concentrations in the model. We will need to conduct these sensitivity simulations for both the 2000 and 2050 atmospheres, since the former atmosphere in the GCIAQ simulation will be driven by emissions and meteorological fields (from the GISS GCM) different than those used so far in our GEOS-CHEM work. Consistency is essential here for diagnosing and interpreting trends.

Our analysis of baseline PM concentrations and visibility in the GCIAQ model results will follow the same approach as in our past work and in the work described in Task 1 of this proposal. We will examine means, frequency distributions, and contributions from different sources. We will quantify trends between 2000 and 2050 and interpret these trends

in terms of the underlying forcing factors. For specific application to the RHR we will compare the 2000 and 2050 values of the 20th percentiles of baseline and natural visibilities at the IMPROVE sites.

Schedule. We will focus Year 1 on tasks 1-2, and Year 2 on tasks 3-4. The 5-year simulation for tasks 2 and 3 will take most of Year 1 to complete, but we will proceed with analysis of results as the simulation progresses.

Budget. We request total funding of 213K for the period 2/15/04-2/15/06 (103K for Year 1, 110K for Year 2). A detailed budget will be provided upon request. Personnel costs include:

- **Daniel J. Jacob** (P.I., 2 weeks summer/yr) will direct the project and will be responsible for its execution and delivery of the products to EPRI. Prof. Jacob's academic-year salary is covered by Harvard University; he will devote substantially more time to the project than is charged to EPRI.
- **Rokjin J. Park** (postdoctoral fellow, 60%) will conduct the simulations, evaluate model results with observations, and interpret the results.
- **Robert M. Yantosca** (software engineer, 10%) will support the model environment and conduct simulations as necessary.

Facilities. All simulations will be conducted on SGI machines belonging to our group and dedicated to GEOS-CHEM simulations. Our budget includes 10K/yr to support the maintenance and management of these machines.

I look forward to hearing from you.

Best Regards,

Daniel J. Jacob

REFERENCES

- Chung, S., and J.H. Seinfeld, Global distribution and climate forcing of carbonaceous aerosols, *J. Geophys. Res.* *107*, 4407, 10.1029/2001JD001397, 2002.
- Duncan, B.N., R.V. Martin, A.C. Staudt, R. Yevich, J.A. Logan, Interannual and Seasonal Variability of Biomass Burning Emissions Constrained by Satellite Observations, *J. Geophys. Res.*, *108*, 4040, 2003.
- EPA, Draft Guidance for Estimating Natural Visibility Conditions under the Regional Haze Rule, U.S. EPA OAQPS report, September 27, 2001.
- Gilliland, A.B., R.L. Dennis, S.J. Roselle, and T.E. Pierce, Seasonal NH₃ emission estimates for the eastern United States based on ammonium wet concentrations and an inverse modeling method, *J. Geophys. Res.*, *108*, 4477, 2003a.

- Gilliland, A.B., H.-K. Im, and M.L. Stein, Inverse modeling to estimate NH₃ emission seasonality and the sensitivity to uncertainty representations, presented at the NARSTO Emission Inventory Workshop, Austin, Texas, October 14-17, 2003.
- Ginoux, P., M. Chin, I. Tegen, J.M. Prospero, B. Holben, O. Dubovik, and S.-J. Lin, Sources and distributions of dust aerosols simulated with the GOCART model, *J. Geophys. Res.*, *106*, 20,255-20,273, 2001.
- Griffin, R.J., D.R. Cocker III, R.C. Flagan, and J.H. Seinfeld, Organic aerosol formation from the oxidation of biogenic hydrocarbons, *J. Geophys. Res.*, *104*, 3555-3567, 1999.
- IPCC 2001, *Climate Change 2001: The Scientific Basis*, J. T. Houghton et al. (eds.), Cambridge University Press, UK. 2001.
- Mari, C., D.J. Jacob, and P. Bechtold, Transport and scavenging of soluble gases in a deep convective cloud, *J. Geophys. Res.*, *105*, 22,255-22,267, 2000.
- Park, R. J., D. J. Jacob, M. Chin and R. V. Martin, Sources of carbonaceous aerosols over the United States and implications for natural visibility, *J. Geophys. Res.*, *108*, 4355, doi:10.1029/2002JD003190, 2003.
- Zender, C.S., H. Bian, and D. Newman, The mineral Dust Entrainment and Deposition (DEAD) model: description and 1990's dust climatology, *J. Geophys. Res.*, *108*, 4416, 2003.