Project Title: **Hybrid Source Apportionment Model: an operational tool to distinguish wildfire emissions from prescribed fire emissions in measurements of $PM_{2.5}$ for use in visibility and PM regulatory programs**

Announcement for Proposals and task statement this proposal is responding to:
Announcement for Proposals, 2005-3: Task 1 Air Quality, especially subtask 3 to develop source apportionment techniques.

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Duration of Project: **3 years**

Abstract:

Air quality regulations have the goal of reducing haze in national parks and wilderness areas to natural conditions, and require that fine particulate matter ($PM_{2.5}$) be reduced below a threshold that adversely impacts health. The federally funded and managed IMPROVE and STN monitoring networks track the progress toward these goals. Carbonaceous material is often the largest contributor to haze and $PM_{2.5}$, and semi-quantitative analyses suggest that smoke from fire-related activities is a significant contributor to this carbonaceous material. Fuel reduction activities mandated by the National Fire Plan along with on-going reductions in non-carbon pollutants will serve to further increase the relative contribution of carbonaceous material to $PM_{2.5}$ and haze. This may lead to demands that managed fire activities reduce pollutant emissions. To develop meaningful control strategies, federal land managers and policy makers need tools to apportion daily measurements of $PM_{2.5}$ at IMPROVE and STN sites to smoke from natural, e.g. wildfire, anthropogenic, e.g. some prescribed fire, and international fires, as well as mobile and industrial sources. Traditionally, source attribution tools use either air quality models, which attempt to simulate the contributions of sources based upon first principles, or source receptor models which apportion measured $PM_{2.5}$ based upon measured marker species and their source profiles. Neither technique is adequate. Air quality models can apportion $PM_{2.5}$ to all source types, but the results often include excessive errors due to limitations in model inputs and chemistry and dispersion mechanisms. Receptor models are constrained by measured data, bounding the estimates, but they cannot apportion mass to different fire types, e.g., wild and prescribed fire, and are limited in apportioning secondary aerosols. We propose to develop a new tool based on a hybrid source apportionment model (HSAM), which incorporates air quality modeling results into a new type of receptor model. HSAM will be capable of apportioning primary and secondary aerosols in measured $PM_{2.5}$ to contributing source types, including different fire types, with associated uncertainties on a daily basis. HSAM will be thoroughly tested using data from the IMPROVE, STN, and other monitoring networks and results from multiple air quality modeling runs to assess HSAM capabilities and uncertainties. HSAM will then be applied to all IMPROVE and STN monitoring sites from 2002. Fully documented HSAM and 2002 apportionment results will be made available via the IMPROVE and VIEWS websites. Last, a workshop will be convened with modeling centers tasked with routine air quality modeling of smoke and $PM_{2.5}$, such as the Forest Service's BlueSky and FCAMMS, to facilitate the incorporation of HSAM into their programs. This project will provide land managers with tools to help them quantify the usefulness of emissions reduction techniques for application in Smoke Management Programs. This will ensure that fire managers are able to maintain the
capability to use prescribed fire and quantify the effectiveness of emissions reduction techniques.

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1 Introduction

1.1 Project Justification

Health and visibility (haze) regulations require reductions in atmospheric fine particulate matter ($PM_{2.5}$). Evidence suggests that carbonaceous material from smoke is a significant contributor to $PM_{2.5}$. With the planned increase in prescribed burning to reduce fuel build up, detailed in the National Fire Plan (NFP), and planned reductions in non-fire related emissions, smoke's contribution to $PM_{2.5}$ and haze will likely increase. In light of this, regulators will likely look to anthropogenic sources of smoke to achieve further decreases in $PM_{2.5}$ and haze. Therefore fire managers, air quality regulators and policy makers need to distinguish what portion of measured fine particulate ($PM_{2.5}$) comes from fire, and, of that amount, to determine how much comes from domestic natural wildfire, from agricultural burning, and from prescribed fire. In addition, they need to quantify the effectiveness of *emissions reduction techniques* they are mandated to apply by *Smoke Management Programs.* The tools required to accomplish this are currently insufficient. This proposal will develop a tool to apportion contributions from a wide variety of all domestic and international sources to measured carbonaceous components of $PM_{2.5}$ and impaired visibility. The apportionment tool will quantify the contributions made by natural and anthropogenic fire types and by international sources to each daily measurement in the IMPROVE and STN regulatory networks.

1.1.1 Regulatory drivers

The Regional Haze Rule (RHR) enforces the goal of the Clean Air Act that haze in our national parks and wilderness areas (class I areas) be returned to natural conditions. The regulation requires that states through Regional Planning Organizations (RPOs) develop emission reduction plans for the particulate matter that causes haze in order to make progress toward achieving natural visibility conditions within the next 60 years. Progress is measured every five years by comparing haze levels for the worst haze days against the goal. It is required that haze remains the same or is reduced compared to the previous five years. To implement the rule, air quality managers need to know the haze level at each class I area and the contributions from all sources including anthropogenic and natural smoke. The Interagency Monitoring of Protected Visual Environments (IMPROVE), a federal land management agency/EPA monitoring program, was established to track progress toward this national goal. However, current monitoring technology is not capable of apportioning or separating anthropogenic emissions such as mobile sources or industry-related activity from wild or prescribed fire emissions. Based on IMPROVE data, all class I areas are exhibiting haze above the officially defined natural background estimate (Copeland, 2004).

The $PM_{2.5}$ National Ambient Air Quality Standards (NAAQS) are designed to protect human health and require that $PM_{2.5}$ concentrations not exceed 65 $\mu g/m^3$ over a twenty-four hour period or 15 $\mu g/m^3$ for an annual average. A number of eastern U.S. and southern California counties do not "attain" this level of air quality and are likely to be designated as $PM_{2.5}$ *non-attainment areas* (http://www.epa.gov/pmdesignations/) By law, *non-attainment areas* will be subject to a number of strict requirements to reduce particles and the pollutants that form from them including the anthropogenic contribution to fire emissions. The Speciated Trend Network (STN) is a U.S. EPA monitoring network, similar to IMPROVE, established to help understand the causes of $PM_{2.5}$ exceedances and trends in $PM_{2.5}$ over time.

1.1.2 How much does fire contribute to regional Haze and $PM_{2.5}$?

Carbonaceous material accounts for 30–50% of $PM_{2.5}$ mass in urban areas (U.S. EPA 2004a) and 30–90% of haze in rural areas (Malm et al., 2004a). As shown in Figure 1, particulate organic carbon accounts for over 50% of the haze in the Northwest and Sierra Mountains on the 20% worst visibility days in 2002. These are the days that the RHR is most concerned about. Recent studies have shown that 80–95% of summertime carbonaceous material in the rural southeast and western U.S. is due to biogenic sources (Lewis et al., 2004; Bench and Herckes, 2004). However, it is not known how much of this organic mass is attributed to uncontrollable sources such as wildfire, biogenic emissions, and international contributions (Ito and Penner, 2004) and how much is due to anthropogenic sources such as agricultural fire, some prescribed fire, and

residential wood combustion.

Some situations illustrate an obvious link to wildfire. Figure 2 presents the particulate organic carbon measured at Monture, Montana, from 2000 to 2003. Over this period there were two episodes (8/00 and 8/03) where 10s of $\mu g/m^3$ of organic carbon were measured. Satellite imagery for these two episodes reveals that the monitoring site was embedded in thick smoke plumes on both occasions. Under these conditions apportionment to wildfire is obvious and can be made with little uncertainty. However, excluding these obvious smoke events, every spring, summer, and fall organic concentrations are often between 1 and 10 $\mu g/m^3$ which is high enough to significantly impair visibility. This raises the question of how much of the $PM_{2.5}$ on these days is due to natural or human-caused sources. A second example from Florida illustrates the aerosol composition on April 8, one of the measured worst 20% visibility days in 2002 at this site (Figure 3). What sources contributed to the 20 $\mu g/m^3$ of organic carbon aerosol measured on this day? An apportionment of $PM_{2.5}$ on these days is exactly what needs to be done by either state regulators or the fire community for both regulatory programs (for example, see the WRAP assessment process at http://www.wrapair.org/forums/aamrf/index.html).

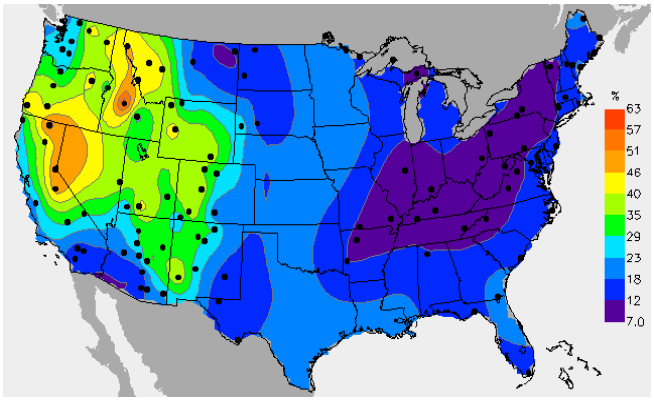

Figure 1. Contribution of organic mass to haze in class I areas on the 20% worst visibility days during 2002.

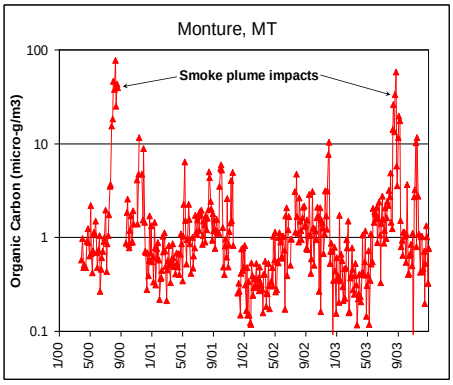

Figure 2. The organic carbon concentrations at Monture Montana in the IMPROVE monitoring network

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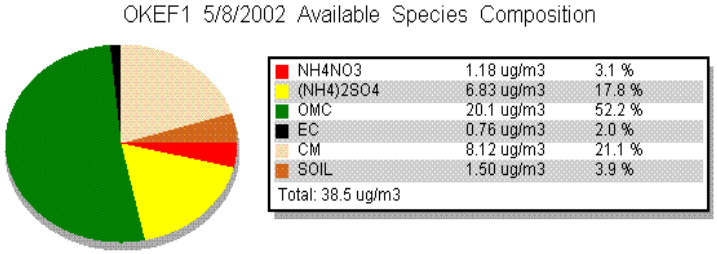

Figure 3. The $PM_{2.5}$ composition at Okefenokee, GA, on one of the highest measured concentration days during 2002.

1.1.3 Implications of regulations for smoke management

States are developing plans to reduce measured haze to natural background and $PM_{2.5}$ concentrations below the health standard. This work includes classifying all burning as either natural or anthropogenic and, for the portion deemed anthropogenic (agricultural burning and certain prescribed fires), mandating the application of *emissions reduction techniques* through *Smoke Management Programs*. Current estimates are that smoke is a significant contributor to haze and $PM_{2.5}$. Without additional information, regulators will look to anthropogenic smoke for reductions. Certainly these regulations have the potential to affect National Fire Plan (NFP) fuel reduction plans (http://www.fireplan.gov/index.html) by limiting the fire managers' ability to reintroduce and increase the use of fire, especially in the urban/wildland interface. Especially as regulatory programs reduce industrial and mobile source emissions (at significant cost and inconvenience to citizens) it becomes increasingly important for the fire community to **quantify their contributions to regional haze and $PM_{2.5}$.** *Smoke Management Programs* will acknowledge that fire can be both natural and anthropogenic but will require the use of apportionment tools to quantify fire's contributions to measured $PM_{2.5}$ (Riebau and Fox, 2001).

1.2 Project Objectives

The primary objective of this project is to **develop an unambiguous and routine methodology and tool for apportioning carbonaceous and other compounds in $PM_{2.5,}$ measured in the IMPROVE and STN networks to smoke from prescribed, wild, agricultural, and transboundary fire activities as well as other sources on a daily and annual basis.** This proposal directly responds to "Task 1-Air Quality" of AFP 2005-3 of the Joint Fire Science Program.

The tool will be based on a new hybrid source apportionment model (HSAM) that uses measured data from the IMPROVE or STN networks and links it with air quality modeling results. The air quality modeling will not be done as part of this project but will come from collaboration with modeling being done by our group (NPS/CIRA), by cooperators at the National Center for Atmospheric Research (NCAR), the Regional Planning Organizations (RPOs), the U.S. Forest Service (FCAMMS) and others. This project will build on the previous and current research in the National Park Service on source apportionment tools and analysis and use of the IMPROVE aerosol data.

The second objective will be to **extensively evaluate HSAM** using several air quality monitoring data sets, including routine data from the IMPROVE network, research data collected as part of special monitoring studies, and results from two air quality modeling studies. Once tested, HSAM will be used to apportionment IMPROVE and STN samples during 2002 to smoke and other source types.

The third objective is to **deliver a proven source apportionment tool** to federal land managers, air quality regulators, and policymakers through written reports, a workshop and training session for federal land managers, regulators, and policymakers, and a series of peer-reviewed articles documenting the development, validation, and application of the source apportionment tool. In this last phase we will work with other organizations developing routine fire emissions and air quality simulations to incorporate their results into the source apportionment tool, allowing the generation of high quality smoke apportionment results on a routine basis.

1.3 Background

Particulate organic matter is a complex mixture of directly emitted particles from different combustion sources and secondary organic aerosols derived from emissions of volatile and semi-volatile organic compounds from both anthropogenic and natural sources. Any effective source apportionment tool needs to be able to separate out the contributions of the different sources and source types to both primary and secondary aerosols. Traditionally, source apportionment is conducted using air quality simulation models and source receptor models. Air quality modeling attempts to directly simulate pollutant emissions, transport, chemical transformations, and physical removal mechanisms using first principles. Source receptor modeling attempts to apportion measured particulate mass to various source types using measured chemical and physical characteristics of the particulate matter. As discussed below, neither technique is adequate to apportion smoke's contribution to $PM_{2.5}$ with sufficient accuracy and temporal resolution or to differentiate between smoke source types, i.e. wild, prescribed, and agricultural fire. We propose to develop a new technique based on hybrid modeling, which combines the benefits of both source and receptor modeling.

This background section reviews the various smoke apportionment techniques, identifying their strengths and limitations. We describe current hybrid modeling research activities by the NPS/CIRA group and we illustrate why hybrid modeling is the most appropriate technique for routine smoke apportionment.

1.3.1 Air quality simulation modeling

Air quality simulation models, in theory, identify and quantify the amount of pollution each source contributes to any receptor or measurement. For this reason, many organizations have pursued the development and use of models for simulating the impact of fires and other sources on haze and $PM_{2.5}$. This includes the RPOs (WRAP, 2004; VISTA, 2004, MRPO, 2004), the U.S. EPA (U.S. EPA, 2004b), the Forest Service, the NPS/CIRA team, and NCAR, among others. However, these models are limited by our understanding of atmospheric processes, our ability to simulate these processes, and the quality of data inputs for the models. For modeling biomass burning, the biggest limitations are estimating smoke emissions,

representing meteorology in complex terrain, and understanding the chemical processes which lead to the formation of secondary organic aerosols (SOA).

Estimating fire emissions of gaseous and particulate products from the many phases of different types of fire, pyrolysis, and gas and solid phase oxidation processes is not only difficult but may be impossible (Andreae and Merlet, 2001; Sandberg et al., 2001). In addition, fuel types and conditions vary so that the prediction of fire evolution is difficult (Graham et. al., 2004). Consequently, there is a poor understanding of particulate and volatile organic carbon (VOC) emissions (Battye and Battye, 2001) and current fire emission inventories lump all VOCs into one category (Houyoux and Vukovich, 1999; FEJF, 2004). As a result of this complexity, fire emissions are generally developed outside the model, often by a laborious hand process. In order to improve fire emissions inputs for air quality simulation models we are, with partial funding from the JFSP, developing a Community Smoke Emissions Model (CSEM) (Barna and Fox, 2003). Appendix 1 describes CSEM and the context for its development. CSEM will be available to supplement fire emissions inventories used in this proposal. Similarly, natural emissions of biogenic volatile organic carbon (BVOC) from vegetation need to be included in air quality simulation models. The complexity and approximations needed to routinely specify them remains a limitation of air quality simulation modeling (Wiedinmyer et al., 2001; Vukovich and Pierce, 2002).

The second source of errors for smoke modeling comes from the meteorological data used to drive regional scale models. Although the Forest Service FCAMMS are in the process of developing the needed higher resolution meteorological inputs, the meteorological data sets used to date for continental scale air quality modeling have generally been resolved on coarse grids on the order of 36 km or greater. This is too coarse to capture much of the import forcing processing that complex terrain has on airmass transport, such as channeling by canyons and upslope flows (Whiteman, 2000). However, many fires occur and many class I areas are located in complex terrain. In addition, current meteorological models do not assimilate measured precipitation and cloud data, leading to large errors (Stauffer and Seaman, 1994). Precipitation and clouds play a vital role in pollutant deposition and secondary formation of organic and other aerosols, and errors in these parameters can cause large errors in air quality modeling. In a recent modeling study, 36 km MM5 wind fields were used to simulate haze in Texas. During July 1999, the MM5 data overestimated the frequency of precipitation by more than a factor of 2 (Barna et al., 2004). During this period, the model severely underestimated measured sulfate concentrations.

The third cause of error is the simulation of secondary organic aerosols (SOA) from organic gases emitted by fires and biogenic sources. The atmospheric reactions of organic gases leading to organic particulates are very complex and not fully understood even in ideal laboratory situations (Seinfeld and Pankow, 2003). For example, the scientific community has only just become aware of the importance of acid aerosols in the catalyst reaction of organic gases to form SOAs (Jang et al., 2002). The current formations of SOA in models are still crude estimations and as a result, considerable error is introduced in modeling results (Pun et al., 2003).

The accumulation of these errors can lead to large modeling errors, reducing the utility of modeling results. Figure 5 presents the latest modeling results from the WRAP RPO of organic carbon (OC) for July 2002 at Yosemite NP, CA, and Spokane WA. At Yosemite NP, the simulation is poor with the model systematically underestimating the measured OC, missing the peak OC concentrations on day 194, and even being anti-correlated during the last three days. On the other hand, at Spokane, WA, the simulation systematically overestimated the OC concentrations but reproduced the temporal variability of the measured data.

It is necessary to use air quality modeling for regulatory applications, but, for the reasons stated here, one can not rely on it entirely to apportion carbon aerosol at measurement points.

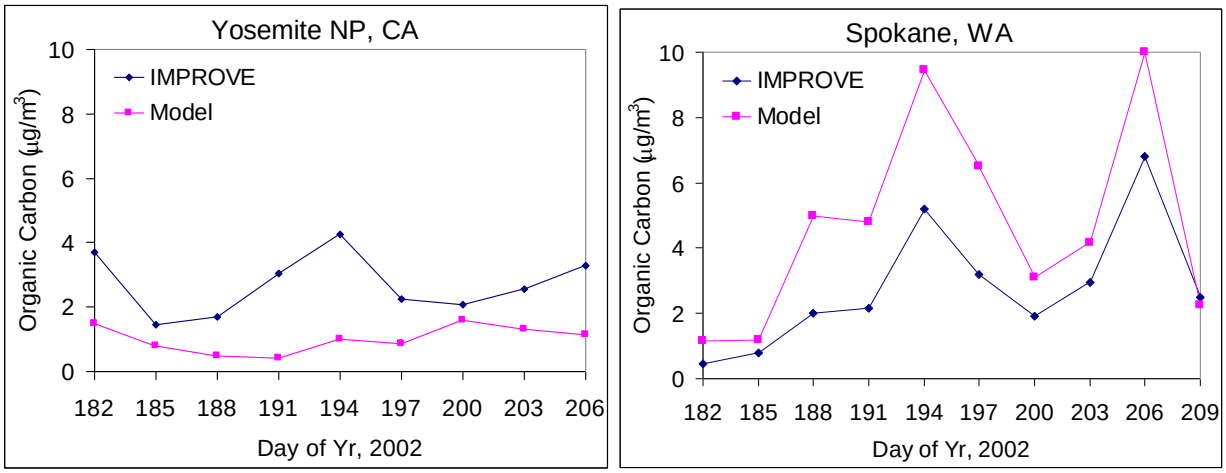

Figure 5. Comparison of CMAQ model organic carbon concentration to measured data at the Yosemite NP, CA, and Spokane WA, IMPROVE monitoring sites during July 2002.

1.3.2 Source receptor modeling

A unique feature of atmospheric aerosols is that they carry their own chemical "fingerprint". For example, the composition of biomass smoke includes organic and elemental carbon as well as numerous trace compounds. On the other hand, the composition of power plant emissions is dominated by sulfur species while soil dust is composed of elements from the earth's crust. These chemical fingerprints or source profiles can be used in receptor models to identify and divide the fine particulate mass measured at a receptor into contributions from smoke, mobile, and other source types.

The most popular and traditional source receptor modeling approach is the chemical mass balance (CMB) model (Gordon, 1980; Watson, 1984). CMB uses known physical and chemical characteristics of aerosols, such as ratios of tracer species, at the receptors and sources to attribute primary aerosols to single sources or source types. The CMB model relies on the assumption that the concentration of airborne aerosol species can be described by the sum of a number (hopefully, small) of source profiles. The equation for this description is:

$$c_{ki} = \sum_j S_{kj}\, a_{ij} + \varepsilon_{ki} \qquad (1)$$

where

k	=	1 ... m, the number of observations,
i	=	1 ... n, the number of marker species,
j	=	1 ... N, the number of sources,
c_{ki}	=	concentrations of aerosol species (including marker species) i for time period k,
S_{kj}	=	relative contribution of source type j to observation k at the receptor, and
a_{ij}	=	source profiles. The relative concentration of species i in source type j
ε_{ki}	=	model residual

The source profiles a_{ij} are the relative contribution of the marker species of the source to the total ambient mass from that source. These are typically determined by measuring fresh emissions from a source and thus representing the primary emitted mass. Important assumptions associated with CMB include: 1) the composition of emissions are known and constant, i.e. a_{ij} is known, and 2) aerosol components do not react with each other, i.e., the tracer ratios associated with a source (e.g., smoke) remain constant over time. Thus, the CMB approach is limited in its ability to resolve secondary species. If these assumptions are violated, the apportionment is degraded (Watson and Robinson, 1984; Malm et al., 1989). In the case of smoke, these conditions are only met near the fire. In aged smoked, half of the $PM_{2.5}$ can be made of secondary products formed by reactions of VOC gases in the atmosphere during travel (Reid and Hobbs, 1998).

For CMB to identify and assess the contribution of smoke to a receptor, unique smoke marker species, characterizing the source profile of the fuels, need to be identified. A number of researchers have performed

detailed organic speciation using techniques such as gas chromatography followed by mass spectrometry (GC/MS) on organic smoke aerosols in the laboratory and field (Schauer, 1998;, Simoneit et al., 1999; Ferek et al., 1998; Zheng et al., 2002). These studies and others have shown that there is no single stable source profile for smoke from biomass burning. The relative concentrations of the marker species vary with the nature of the fire, e.g., a flaming or smoldering fire and the fuels consumed. For example, levoglucosan is one of the most promising smoke tracer species (Simoneit et al., 1999), but the relative concentration of levoglucosan in smoke can vary over an order of magnitude depending on the fire and the age of the smoke when sampled.

Using the smoke organic marker species, their source profiles and the CMB model, the contribution of primary smoke particles to $PM_{2.5}$ has been determined in a number of studies. For example, Zheng et al. (2002) and Schauer and Cass (2000) used CMB and organic source profiles to apportion fine particulate matter and organics to various source types including wood smoke in the southeastern United States and southern California. An interesting aspect of Schauer and Cass' (2000) work is that they were able to identify unique marker species for soft- and hardwoods and used these to separate their contributions to the measured carbonaceous material. However, they could not apportion secondary material which accounted for up to 80% of the organic mass during one episode studied.

CMB modeling can be used to apportion primary aerosol species on a sampling-period-by-sampling-period basis. However, if the data set contains an adequate number of samples, multivariate algorithms can be used to derive source profiles, $\boldsymbol{a}$, and source contributions, $\boldsymbol{S}$, using only measured aerosol composition. These techniques are based on regression and factor analyses (Hopke, 1985; Malm et al., 1989). Since the source profiles are derived from the data, it is possible to apportion secondary and primary aerosols. For example, Malm and Gebhart (1997) showed that under limited transport regimes using a modified CMB approach along with linear regression analysis, about 50% of sulfate measured at Grand Canyon during the winter months was attributed to smoke. Two new receptor-oriented multivariate models are the Positive Matrix Factorization, PMF (Paatero, 1998), and UNMIX (Henry, 2000). Several studies have successfully used these tools and data from the IMPROVE monitoring network, or similar data, to apportion measured $PM_{2.5}$ to biomass burning smoke and other sources (Poirot et al., 2001; Kim and Hopke, 2004). These studies illustrate that, using only routinely available IMPROVE data, it is possible to separate out the average smoke contribution to measured $PM_{2.5}$ over an extended period of time.

Source receptor modeling is a powerful technique for identifying the contribution of smoke and other source types to the measured $PM_{2.5}$. However, the techniques are limited and by themselves are not adequate for assessing smoke's contribution to haze and $PM_{2.5}$. CMB can apportion smoke to measured mass on a daily basis, which is needed for the regional haze rule, but requires detailed speciation of the carbonaceous aerosol component, which is expensive and not available from existing routine monitoring networks. CMB also cannot apportion secondary organics, which could account for half of smoke mass (Reid and Hobbs, 1998). Factor and regression analysis techniques, can apportion the secondary smoke contribution using routinely available data from the IMPROVE and STN networks, but are unable to provide the needed time resolution. In addition, no receptor model alone can distinguish between wild and prescribed fires. Finally, CMB requires that source profiles are fixed *a-priori*, but in reality these profiles will vary based on fuel, fire characteristics, and the age of the smoke when sampled.

1.3.3 Hybrid source apportionment modeling

Air quality simulation and source receptor models each have different strengths and weaknesses. Air quality simulation models can apportion secondary and primary species and the identify contributions from individual fires, thus allowing the separation of wild, prescribed, and agricultural fires. However, the information requirement and chemical mechanism of these models are incomplete and the results are not constrained by measurements, leading to large errors and biases. On the other hand, the source receptor models are constrained by measured data, bounding their errors, but apportioning secondary particulate matter and between smoke types is problematic. Therefore neither technique adequately addresses the needs of the air quality regulations for smoke apportionment.

Hybrid source apportionment modeling directly combines measured data with air quality modeling

results, ideally preserving the temporal and source type resolving power of the air quality model, but has results that are bounded by the measured data and satisfy the source profiles. Hybrid models can be formulated to directly incorporate measured data into an air quality model using data assimilation and model inversion techniques. A number of researchers have done this in order to derive or improve upon emission inventories (e.g., Mendoza-Dominguez and Russell, 2001; Enting, 2002). These are powerful techniques, but require extensive model development and do not incorporate many of the measured tracer species, e.g., potassium and bromine, used in receptor models since air quality models do not simulate them.

The NPS/CIRA group has pursued an alternative approach of incorporating airmass dispersion and air quality modeling results into a source-receptor oriented framework. Two methods pioneered by this group are the trajectory mass balance (TrMB) and forward mass balance regression (FMBR). These transport receptor models develop statistical relationships between airmasses transported from the source regions to receptor areas and measured data at the receptor. These relationships allow the apportioning of measured mass to selected source regions. Schichtel et al. (2004) derive the theoretical basis for these models, fully evaluate them, and apply them to apportion particulate sulfate concentrations at Big Bend NP, Texas, to different source regions in North America.

Ames et al. (2004) applied the TrMB method in conjunction with a wildfire occurrence and acreage-burned data set to apportion organic mass measured by the IMPROVE program to wildfire, both primary and secondary contributions (see Appendix 1 for a discussion of this approach and others). Figure 5 presents the annual apportionment of wildfire to IMPROVE organic mass for 2000 and 2001. During 2000, large wildfires in Idaho, Montana, and Wyoming were found to be responsible for over 75% of the organic mass. However, in 2001 less than 50% of the organics were due to wildland fire smoke. This technique can also be used to apportion the contribution of prescribed and agricultural fires to IMPROVE organic concentrations, provided their emission inventories exist.

TrMB and FMBR can only apportion average contributions over long time periods, on the order of a season to year. To obtain higher time resolution, results from air quality simulation models need to be used. Schichtel et al. (2004) merged CMAQ source apportionment modeling results and measured particulate sulfate concentrations using a synthesis inversion methodology (Enting, 2002) to apportion particulate sulfate at Big Bend NP, Texas. This allowed for the apportionment of particulate sulfate at Big Bend to North American source regions on a near daily basis. In the analysis, the hybrid technique significantly changed and improved the initial CMAQ air quality modeling source apportionment results. This is significant because, CMAQ is the current state of the art air quality simulation model and sulfates are among the best understood aerosol components, yet still the modeling results alone were inadequate to apportion the particulate sulfate. A shortcoming of the synthesis inversion approach is that again the tracer elemental and other marker species were not incorporated into the method. Therefore not all of the available information was used.

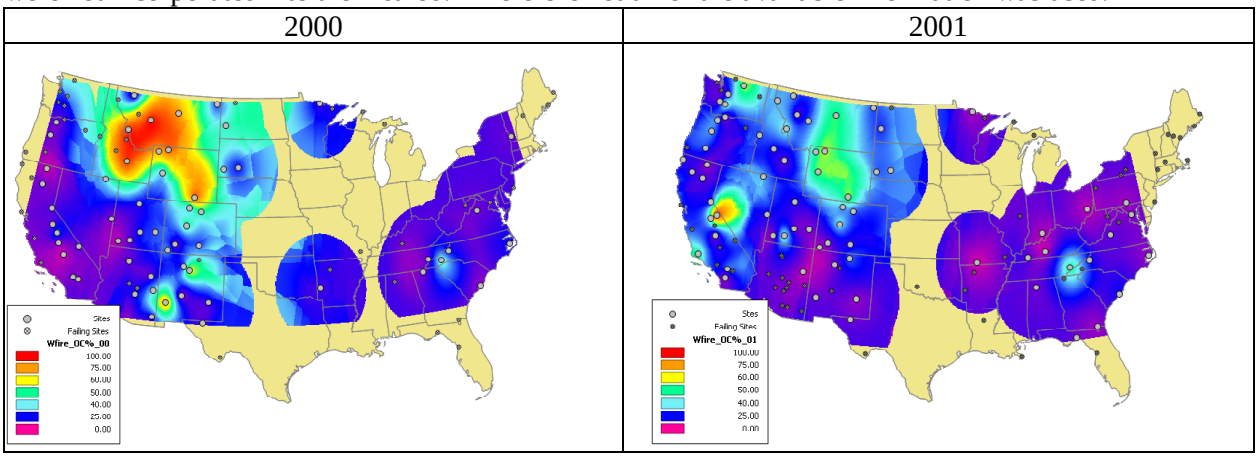

Figure 5. The annual relative contribution of smoke to IMPROVE organic mass for 2000 and 2001.

2 Materials and Methods

In the proposed project, we will expand on air quality simulation and source-receptor and hybrid

modeling techniques that have been developed and applied by the NPS/CIRA group. The smoke apportionment tool we are proposing will be a receptor oriented hybrid model that incorporates all available information including air quality model source apportionment results and source marker species measured at a receptor. The technique will allow for the apportionment of both primary and secondary aerosol species and separation of smoke from wild, prescribed, agricultural, and trans-boundary fires as well as other sources.

This section describes the proposed hybrid model for estimating source contributions and their uncertainties to measured $PM_{2.5}$. In addition, the inputs to the hybrid model and proposed testing and evaluation of the tool are discussed.

2.1 Hybrid Source Attribution Model (HSAM)

The new hybrid model will be based upon the CMB model in equation 1. In the CMB model it is assumed that the source profiles are known with no error and the measured concentrations are known with error, $uc_{k,i}$. The CMB equation is then solved for the source contributions $\boldsymbol{S}$. This is typically done using a least square regression technique, which uses the squared residual as a measure of the quality of model fit and solution. The optimal solution is then the $\boldsymbol{S}$ that minimizes the sum of all squared residuals:

$$Qc = \sum \frac{(c - \hat{c})^2}{uc^2} \tag{2}$$

where c is the measured concentration, $\hat{c}$ is the estimated concentration from $\boldsymbol{S}$ and $\boldsymbol{a}$, and uc is the uncertainty or standard deviation associated with c.

The least square CMB model can be generalized to include air quality source apportionment results for some or all sources and source types. In this case, the minimization term Q would contain another measure of the quality of fit accounting for the deviation from the *a-priori* source apportionment results:

$$Qs = \sum \frac{(S_m - \hat{S})^2}{uS_m^2} \tag{3}$$

where S_m is an air quality model estimate for an element of the source contribution matrix $\boldsymbol{S}$; $\hat{S}$ is the corresponding fitted value, and uS_m is the uncertainty estimate for S_m, or a measure of the difference between the numerical model estimate and true source contributions. The sum is taken over all the elements of the $\boldsymbol{S}$ matrix.

As discussed previously, the source profiles for smoke can vary substantially based on fuels, fire conditions, and the production of secondary species during aging. Therefore a third term is added to the optimization which allows for the source profiles $\boldsymbol{a}$ to vary within a predetermined range:

$$Qa = \sum \frac{(a - \hat{a})^2}{ua^2} \tag{4}$$

where a represents an *a-priori* source profile value and $\hat{a}$ the corresponding fitted value. The quantity ua is the uncertainty associated with our initial specification of a. The sum is over all sources and all species.

Ideally, all three "discrepancy measures" or "quality of fit" measures should be small. However, in practice what we seek is a model fit for which Qc is acceptably small while Qa, and Qs are also small. There are at least two ways to formulate these requirements. One approach consists of defining an objective function Q which is an appropriately weighted average of these individual discrepancies. We would then find a model fit for which Q is the smallest. Another approach is to minimize Qc, but require that Qa, and Qs are all smaller than some prescribed threshold values. Regardless of the approach taken, the model parameters will be required to be positive to make them physically interpretable. In either case the optimization problem involves minimization of a nonlinear objective function with quadratic constraints. A number of algorithms are available for carrying out the constrained optimization using variations of the Newton-Raphson method. For instance, see Schnabel and Dennis (1983).

Uncertainties in the predictions made using the fitted model can be assessed using cross-validation methods whereby the full data set is split into a "training data set" and a "test data set". The model is fitted

using the training data set and the quality of the fit is evaluated by predicting the "test data set" using the model and comparing the predictions to the known values. Cross validation will aid in the derivation of realistic uncertainties without relying on distributional assumptions, which are often made out of convenience and not necessarily to reflect reality.

This hybrid model can be applied to a single sample or multiple samples. However, it is desirable to incorporate multiple samples to further constrain the optimization problem, reducing the uncertainties. The problem with simply incorporating multiple samples is that the source attribution is truly valid for only the average across all samples used in the analysis. This is because, in the solution of the system of equations for the source contributions, $\hat{S}$, we derive only one source profile matrix $\hat{a}$. This issue can be minimized by incorporating samples that are impacted by a similar mix of sources; therefore the derived source profile matrix $\hat{a}$ would be applicable to individual sample used in the analysis. The most straightforward way to do this would be to use the model results to classify and group the samples.

2.2 Hybrid Source Attribution Model Inputs

The HSAM tool requires three sets of inputs: speciated aerosol concentration data, source apportionment estimates from an air quality model, and initial source profiles. In addition, associated uncertainties are needed for each of these inputs. Speciated aerosol data and their uncertainties are readily available from routine monitoring networks such as IMPROVE and STN which are archived at the IMPROVE and VIEWS websites maintained by NPS/CIRA. These networks do not speciate particulate carbonaceous material, so they contain only a few smoke marker elements. There is promising new research that suggests that inexpensive techniques can be developed to measure levoglucosan (Collett et al., 2004), one of the most promising smoke marker species (Simoneit et al., 1999). Organic smoke marker species have been measured in a number of special monitoring studies, such as in the SEARCH network (Zheng et al., 2002) and Yosemite smoke study (Malm et al., 2004b) which are available for testing the *hybrid modeling* methodology. Measurement errors will be used, but the errors for values near the instrument detection limits are underestimated and special attention will be given to properly estimate the errors of these values.

Smoke source profiles have been developed by sampling and analysis of primary smoke in laboratory settings and from sampling known smoke plumes. A thorough review of this literature will be conducted to develop initial source profiles and uncertainties of these source profiles for HSAM. In addition, if a companion proposal to develop smoke marker measurement methods and source profiles (Malm, 2004) is funded, the new profiles will be used extensively in this work.

A key component of this project is obtaining air quality simulation modeling estimates of prescribed and wildfire contributions to particulate mass. In support of the regional haze regulations, modeling is currently being done by the Regional Planning Organizations (RPOs) (WRAP, 2004; VISTA, 2004, MRPO, 2004), in support of the $PM_{2.5}$ standards by the U.S. EPA (U.S. EPA, 2004b), and for a variety of other purposes by the Forest Service, the NPS/CIRA team, and NCAR, among others. These efforts all use one of three air quality models, Community Multi-scale Air Quality Model (CMAQ) (Byun and Ching, 1999), CAMx (CAMx, 2004), and REMSAD (SAI, 2002) and are driven by regional meteorology, typically at 36 km grid scales. Detailed emission inputs are required including fire and biogenics, which both have varying degrees of accuracy and complexity. The USDA Forest Service is in the process of developing regional air quality simulation modeling capabilities at their Fire Consortia for Advanced Modeling of Meteorology and Smoke (FCAMMS http://www.fs.fed.us/FCAMMS) using CMAQ and more specialized regional models. Modeling individual and multiple fires is also currently being done in a number of locations, most importantly by the BlueSky program (http://www.blueskyrains.org). BlueSky tracks plumes from prescribed and wildfires to estimate their combined downwind ground level concentration. This system is will be operating nationwide (FCAMMS) by 2005, providing a source of specific fire contributions for all modeled fires. Our research team (NPS/CIRA) is implementing the CAMx model to produce national air quality simulations to help interpret IMPROVE data.

The model uncertainty estimates are critical for defining the degree to which the final results should be constrained to the model estimates. Poor modeling results, e.g., organic carbon at Yosemite NP in Figure 5,

need to have large uncertainties to minimize the model's influence on the final results. Modeling results such as for Spokane, WA (Figure 5), contain meaningful information and their uncertainties are smaller, but still large enough to account for any systematic bias in model results. Unfortunately, the development of meaningful uncertainty estimates is rarely part of any modeling study. Therefore we will develop model uncertainties through a detailed evaluation comparing the air quality model results to measured data. One potential method is to use the synthesis inversion technique employed by Schichtel et al. (2004) which develops uncertainty estimates when fitting the model source attribution results to measured data. This method and others will be explored in this project. A major effort of the first year of this project will be devoted to developing meaningful model uncertainty estimates.

2.3 Hybrid Model Testing and Application

After the hybrid receptor model has been developed, it will be tested and evaluated using two modeling and three monitoring data sets. The system first will be tested on speciated aerosol data collected at Yosemite National Park during the summer of 2002. During this time period the National Park Service conducted an extensive aerosol monitoring study (Malm et al., 2004b) collecting daily and hourly speciated aerosol data. In addition, unique measurements were conducted including speciated fine organic particulates and carbon 14/12 analysis which distinguishes between particulate carbon from biogenic and fossil fuel sources. The intent of the project was to study the physical, chemical, and optical properties of carbonaceous aerosols, particularly smoke-related aerosols. Fortunately, during the study the monitoring site was impacted by smoke from local fires in the park; smoke from nearby fires in the Sierra Mountains, and distant fires located in southeast Oregon (Cisneros, 2004). The extensive measurement of smoke from a diverse set of fires provides an ideal opportunity to test the source attribution technique with varying data inputs. For example, this provides the opportunity to test the benefits of using organic tracer species such as levoglucosan in addition to the routine IMPROVE measurements.

Smoke apportionment estimates from air quality models will come from two sources. First, and most important, will be from the air quality modeling conducted by the RPOs. The RPOs are using 2002 as a baseline and their modeling will be completed before the end of 2005. Initial results are already available. The RPOs are primarily using the CMAQ air quality model, driven by simulated meteorology from the MM5 model. RPO emission inventories explicitly include estimates of the emissions from wildfire, prescribed fire, and agricultural burning on at least a daily basis with a spatial resolution appropriate to the grid resolution of the regional air quality simulation. All of the RPO modeling inventories are also using a biogenic emissions model to estimate biogenic gas emissions from vegetation. Thus, all the source types, fire in different forms, biogenic emissions, and of course, industrial and mobile sources are included in the inventory. In addition, the WRAP RPO has spent significant effort identifying four different fire inventories, namely wildfire, "prescribed natural fire", "prescribed anthropogenic fire", and agricultural burning in the western U.S. The RPO modeling results will be available to us (see attached letter from Tom Moore, WRAP technical coordinator) to use in the development of the hybrid model.

A second set of modeling, conducted by our NPS/CIRA team, will be used. NPS/CIRA is conducting regional air quality modeling for the United States at a 36 km scale using the CAMx air quality model, driven by MM5 meteorology. We will utilize RPO fire emissions inventories including, if needed, supplementing them with more explicit fire emissions using CSEM among other tools (Barna and Fox, 2003; Appendix 1).

We will also explore the possibility of using modeling results driven by satellite-derived fire emissions that are being developed and applied by the NCAR Atmospheric Chemistry group (Dr. Christine Wiedinmyer). The NCAR group is conducting regional air quality modeling including fire emissions but focusing on improving biogenic emissions and secondary aerosol formation (Wiedinmyer et al., 2001). A detailed biogenic emissions inventory, including speciated monoterpenes and sesquiterpenes which can produce significant SOA, will be included in these model simulations. Additionally, they have developed an independent fire emissions inventory for all of North America based on the use of MODIS satellite observations of fires at a 1 km spatial resolution. Initial model simulations will be performed for periods in 2001 and 2002 as part of an U.S.EPA STAR grant (PI: Jana Milford, Univ. of Colorado). These simulations have the advantage of an improved biogenic emission inventory and the use of a different fire emission inventory compared to the 2002 WRAP

inventory.

Once the testing and evaluation of the source apportionment technique is completed, the HSAM tool will be applied to the routine IMPROVE and STN data collected during 2002 in the United States. In this analysis, modeling results from both the RPOs and NPS/CIRA will be used. The two source attribution results will be compared to each other. In situations where the two results differ significantly, a judgment on the most correct results will be given. This judgment will be based on the results of the evaluation of the HSAM model and uncertainties derived for the two modeling results.

The measure of success for this project will be whether the HSAM tool can meaningfully separate smoke from wild, prescribed, agricultural, and international fires from each other as well as other sources of carbonaceous material and $PM_{2.5}$. It is difficult to definitively answer this question, since no *a-priori* estimate of the various contributors to haze and $PM_{2.5}$ exist. However, by applying the HSAM tool to multiple ambient and modeling data sets, the range of results can be evaluated providing an assessment of the abilities, uncertainties, and robustness of the HSAM tool. Therefore, when this tool is delivered to air quality managers and policy makers, not only will it provide an estimate of the contributions from various sources, but also an assessment of the uncertainty and quality of these results.

3 Science Delivery and Application

The goal of this project is to develop a hybrid source apportionment modeling (HSAM) tool capable of apportioning contributions of smoke from different types of fire, e.g., wildfire, prescribed fire, agricultural fire, and transboundary fire, and of non-smoke sources to haze and $PM_{2.5}$. In addition, the HSAM tool will be thoroughly evaluated, documented and applied to 2002 IMPROVE and STN data. An important aspect of this project is the dissemination of the HSAM tool and the 2002 source attribution results to user communities. Also important is the adoption of the tool by resource managers, air quality regulators, and policy makers for routine analysis of monitoring data. To increase the chances of success, we will disseminate the HSAM tool and its application results via several avenues:

- written reports documenting the development and validation of the HSAM tool;
- written reports documenting the 2002 source attribution results;
- addition of the HSAM tool, and its documentation, instructions, and illustrations on its use to the IMPROVE/VIEWS websites for public dissemination. In addition, the 2002 results will be incorporated into these websites, leveraging their data visualization and dissemination tools to present the results in various formats suitable for a range of audiences.
- presentations at scientific conferences;
- publication of peer-reviewed journal articles.

Continuing application of the HSAM tool requires the three sets of inputs be provided annually on a continuing basis: measured air quality data; emission source profiles; and air quality simulation model results. IMPROVE and STN data are readily available and source profiles will be developed as part of this (and other) project(s), leaving source attribution estimates from air quality simulation models as the one HSAM component not readily available. Therefore, for this tool to be used in a routine manner, it needs to be incorporated into a modeling center performing routine smoke and $PM_{2.5}$ modeling. To facilitate this, we will convene a workshop of organizations operating air quality models on a continuing and regular basis. This includes the Forest Service through their FCAMMS/BlueSky program, the U.S. EPA, the Regional Planning Organizations, and state agencies such as the California Air Resources Board (CARB). We have already contacted FCAMMS leadership and RPO technical coordinators receiving expressions of strong interest in this project and its results (see attached letters of support). An important part of the workshop will be to identify the needs of the air quality simulation modelers so that we can modify the HSAM tool to seamlessly be incorporated into their modeling systems. This will minimize the work that a modeling center needs to do to incorporate this tool, thus facilitating its adoption. Facilitating adoption of the HSAM tool into modeling centers is a critical component of this project and we will devote much of the final year to this activity.

4 Deliverables

Deliverable	Description	Delivery Date
Annual reports	Annual report	Feb 15, 2006 Feb 15, 2007 Feb. 15, 2008
Final report	Project final report, summarizing results and deliverables	July 1, 2008
Workshop/training on HSAM	Workshop and apportionment tool training session for air quality modeling centers, air regulators and fire planners and managers.	May 30, 2008
Web site	Smoke apportionment results from 2002 will be included on VIEWS web site	March 1, 2008
Journal articles	Peer-reviewed journal articles describing: (1) Hybrid source apportionment model description; (2) air quality source modeling; (3) application of the hybrid smoke source apportionment tool	Manuscript submission dates: (1) 12/2006; (2) 7/2007; (3) 7/2008

5 Project Timeline

July 2005	Study begins
July 2005-July 2006	Development of hybrid source apportionment tool & completion of air quality modeling for 2002
Feb 15, 2006	Submission of annual report (date per JFSP specification)
May-Dec 2006	Draft manuscript describing apportionment tool
Feb 15, 2007	Submission of annual report (date per JFSP specification)
July 2007	Draft manuscript reporting results of simulation modeling
Mar 2007 – Nov 2007	Application of apportionment tools to 2002 data
Jan 2008	Draft Manuscript on results of 2002 hybrid apportionment tool
Feb. 2008	Hybrid apportionment delivered on VIEWS web service
Feb 15, 2008	Submission of annual report (date per JFSP specification)
Feb 2008 – July 2008 (some activities beginning earlier)	Dissemination of results through written reports, web site, workshops, conference presentations, and peer-reviewed journal articles
May 2008	Modeling Center Workshop/Training
July 2008	Study ends, final report submitted

6 Expected Benefits and Measures of Success

Application of the HSAM tool will ensure that emissions from fire, both natural and human caused, will be properly accounted for in the regulatory process and in the eyes of the public. It will facilitate the development, implementation, and evaluation of needed emission reduction techniques in Smoke Management Programs, but will also provide substantive and quantified data about the relative contributions from natural wildfires, from fires outside the United States, and from agricultural burning. This accounting will help to place smoke resulting from implementation of the aggressive hazardous fuel reduction goals stated in the National Fire Plan in its proper context.

7 Key Personnel

This project will be led by Dr. Schichtel with assistance from Dr. Fox. Dr. Iyer will provide technical assistance on the development of the new hybrid modeling technique. Dr. Barna will coordinate modeling activities at NPS/CIRA with assistance from Dr. Rodriguez. Dr. Wiedinmyer will coordinate interactions with NCAR. Dr. Fox in his role as a national consultant to the USDA FCAMMS will oversee interactions with the Forest Service. A graduate student will be supported to assist in all aspects of the study.

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