Contribution of Smoke Emissions to Secondary Organic Aerosols (SOA): Real-World Evaluation of Fire SOA Emissions Factors from Fires in a Data Management System (REFERS-DMS)

February 20, 2015 revision

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Abstract

The 3-year REFERS-DMS project will advance the fire and air quality communities' understanding of Secondary Organic Aerosols (SOAs) from fires, by systematically testing current fire SOA precursor emission factors (EFs) with the SCICHEM photochemical puff model (PPM) and CAMx photochemical grid model (PGM) and evaluating current and emerging fire SOA precursor EFs using real-world atmospheric chemistry measurements. We do not propose to conduct experimental laboratory or field studies to develop new SOA precursor emission factors. Instead, we will apply and evaluate existing and emerging SOA precursor emission factors (including those developed in other studies under this FON) by methodically cataloging them in a data management system and by using tools routinely applied to assess air quality impacts, ambient measurements, and photochemical modeling. Our proposed approach would address portions of each of the four research questions in the "Contribution of smoke emissions to secondary organic aerosols" FON topic. A fire SOA Testing and Evaluation Toolbox (Toolbox) will be developed using state-of-science fire emissions and PGM/PPM modeling tools and detailed atmospheric chemistry data of observed fire plumes from 2013/2014 field study campaigns. The Toolbox will be used to evaluate fire SOA from other studies, such as SOA precursor emissions factors from smog chamber data, laboratory, and controlled field burning EF studies; SOA production from PGMs/PPMs; and atmospheric field study measurements. The Toolbox will also serve as an expandable, coherent, well-documented library of fire EFs to support the continuing evolution and improvements in EF research.

To robustly examine the JFSP's "SOA precursor emissions factors" and for the sake of clarity, we define 3 terms related to SOA: (1) SOA_{pf} - the emission rates of organic gases, semi-volatiles, and particles that are SOA precursors measured at the tip of a flame in a smog chamber or controlled field experiment (e.g., before significant dilution and mixing with other gases and aerosols occurs in the broader ambient environment); (2) SOA_{aqm} – processed and standardized concentrations of SOA components through an emission and PGM/PPM modeling process; and (3) SOA_{obs} – ambient measurement estimates of SOA on filter media or continuous methods where the measurement represents the integration of emission and atmospheric processes. Our proposal leverages SOA_{pf} development work by others and applies consistent and reproducible SOA_{aqm} and SOA_{obs} methods, and explicitly does not reinvent the work of others. This includes building on the methodology of, for example, Yokelson et al. 2013, May et al. 2014, and Urbanksi et al. 2014 to synthesize a wide variety of emission factor research and combine laboratory and field studies. In addition, we will collaborate closely with current JFSP-funded projects studying phase-partitioning of SOA_{pf} and the modeling processes underlying SOA_{aqm}. The REFERS-DMS study will build off the JFSP Particulate Matter Deterministic & Empirical Tagging and Assessment of Impacts on Levels (PMDETAIL) study that is developing improved SOA_{pf}, fire emissions models and representation of fire SOA chemistry in PGMs, and will use the same core participants. PMDETAIL is performing PGM particulate matter (including SOA_{aqm}) source apportionment modeling of fire emissions for the 2008 and 2011 annual periods and evaluating the PGMs against Organic Aerosol and other measurements including additional filter sampling of key fire marker species (e.g., levoglucosan). We will include the PMDETAIL 2011 modeling with the modeling of the 2013/2014 field study periods, and in this project extend the

existing fire climatologies and emissions inventories within the Toolbox. Although numerous fire SOA_{pf} have been developed under laboratory and controlled field conditions (e.g., Yokelson et al., 2013), there has been little attempt to comprehensively evaluate them using real-world atmospheric measurements. Under REFERS-DMS, a White Paper would first be prepared and subjected to external review, that describes atmospheric SOA processes, the current state-of-science treatments of SOA chemistry in atmospheric models, what types of SOA precursor fire emissions they need and describe fire emissions modeling approaches to support the PGM SOA modules and the JFSP project objectives. There are currently two main different types of treatment of SOA in atmospheric chemistry models (e.g., the CMAQ and CAMx PGMs and SCICHEM PPM): (1) the traditional approach where specific VOC species are oxidized from a Condensable Gas (CG) and SOA pairs that are in thermodynamic equilibrium (Pankow, 1994a,b); and (2) the Volatility Basis Set (VBS) approach which groups organic compounds into a set of volatility bins (basis sets) and shifts their mass between the volatility bins as the SOA ages to describe volatility changes due to atmospheric oxidation and evaporation (Donahue et al., 2006; Robinson et al., 2007). Note that the oxidation of SOA precursors can occur in both gas-phase and aqueous-phase chemistry, although the importance of aqueous-phase chemistry in the oxidation of SOA precursors from fires is not well understood. The White Paper will describe the emissions needs for fire SOA precursors by the various PGM/PPM SOA modules and will be a resource for other researchers who are developing SOA_{pf} whose results can subsequently be evaluated using the Toolbox.

For building the Toolbox to evaluate current and emerging fire SOA_{pf}, REFERS-DMS will take advantage of recent atmospheric field study measurements from the Southeast Nexus (SENEX[1]) for the southeastern U.S. and Studies of Emissions and Atmospheric Composition, Clouds and Climate Coupling by Regional Surveys (SEAC4RS[2]) that collected advanced measurements from aircraft platforms that included plumes from fires in Yosemite and Idaho in 2013. The Summer 2014 FRAPPE: Front Range Air Pollution and Photochemical Experiment (FRAPPE[3]) has been completed and we will work with the FRAPPE Science Team currently working on data analysis to extract episode data with transported fire plume measurements that can be applied through our Toolbox. The 2011 fire emissions and PGM modeling platform developed as part of PMDETAIL, that can differentiate fire emissions by intensity (e.g., flaming versus smoldering), biomass type (10 classifications) and fire type (i.e., wildfire, prescribed burns and agricultural burning), will be used as part of the evaluation toolbox. The SENEX/SEAC4RS/FRAPPE 2013/2014 aircraft field study measurements will be analyzed to identify occurrence of impacts of plumes from fires. The SCICHEM PPM will be used initially to simulate the SOA and other concentrations along the aircraft traverse due to the emissions from fires for all of the identified fire impact events. SCICHEM has many of the same full chemistry algorithms as the PGMs (e.g., CAMx and CMAQ) only uses a Gaussian puff formulation. Consequently, it is more flexible on the wind and dispersion inputs than a PGM. We will use observed fire markers along the aircraft traverse and the SCICHEM PPM transport and dispersion will be defined to match the observed fire plume along the aircraft traverse. SCICHEM can then be applied using various fire SOA_{pf} EFs and the SCICHEM full chemistry algorithms can perform the photochemical and aerosol thermodynamic processing of the fire SOA_{pf} emissions to produce particle SOA_{aqm} and other fire concentration species for comparisons against the aircraft measurements.. These advanced measurements include aerosol mass spectrometry (AMS) measurements that have the capability of distinguishing Primary Organic Aerosol (POA) from SOA_{obs} that routine measurements (e.g., IMPROVE and CSN) fail to differentiate. For several of the most promising aircraft field study fire plume impact events, the CAMx PGM will also be setup and applied to evaluate alternative fire SOApf using the aircraft traverse measurements as well as measurements from ground-based monitors, including IMPROVE and CSN routine observations throughout the U.S. and the SEARCH[4] network in the Southeast and the SOAS Centreville, AL ground-based station.

The Toolbox would then be used to evaluate existing SOA_{pf} and PGM/PPM chemistry modules, including those developed under PMDETAIL for the standard (CG/SOA pair) and VBS SOA_{aqm} modules. We will also work with fire SOA_{pf} developers to evaluate new fire EFs as they are developed and hold workshops/webinars where the needs of atmospheric SOA_{obs} and results of the SOA_{aqm}/ SOA_{pf} evaluation can be discussed and approaches refined. These other fire SOA_{pf} researchers include the group at Carnegie Mellon University (CMU), who are advisors in REFERS-DMS, the NOAA group that plan a trip to the fire laboratory in Summer 2015 to investigate fire SOA precursor EFs, and Dr. Kreidenweis and Dr. Barsanti, whose projects were recently funded by JFSP under this same FON. The Toolbox will carry factors through modeling for comparing to field study's SOA_{obs}. Note that the suite of advanced measurements

[1] http://www.esrl.noaa.gov/csd/projects/senex/
[2] https://espo.nasa.gov/missions/seac4rs
[3] http://www2.acd.ucar.edu/frappe
[4] http://www.atmospheric-research.com/studies/SEARCH/

collected during SENEX/SEAC4RS/FRAPPE will not only allow the evaluation of SOA_{pf}, but can also address the evaluation of fire EFs for other species as well. The Toolbox will not be limited to SOA_{pf} since most lab and field studies measure many major pollutant species (e.g., CO, NO_x, NH_3) even if particularly focused on a subset of trace gases. In addition, certain EFs, such as total $PM_{2.5}$ are important for SOA_{pf} evaluation and maintaining consistency (e.g. mass balance) when choosing a "set" for developing a fire emissions model.

Tools for on-going analyses beyond project life				*Some continued functionality*	*Tools/data during project*	
Upload →	Tag →	Summarize →	Inventory →	Evaluate →	Link →	Refine
SOA_{pf} from current studies, future researchers	Metadata based on fire characteristics, fuels, combustion phase	Create visualizations to organize, compare, analyze SOA_{pf}	Build/export fire EIs; use chosen sets of SOA_{pf} for evaluation	Tools that reveal relationships between SOA_{pf}, fire events/ characteristics, fuels, etc.	Link SOA_{pf}, inventories to PGM model output & direct SOA_{obs}; evaluate uncertainties	Use SOA_{aqm} performance and other tools to answer questions about SOA_{pf}, PGM model processes, fire emissions modeling

The results of new fire SOA EFs and evaluation of other emerging EFs will be integrated using the existing WRAPTools web system[5] as a clearinghouse in which new fire SOA_{pf} can be reviewed and tested. The WRAPTools web system provides a solid basis for visualization and analysis of data, based on WRAP FETS[6] and the JFSP-funded Deterministic and Empirical Assessment of Smoke's Contribution to Ozone[7] ($DEASCO_3$) and PMDETAIL studies. WRAPTools will be extended to include development of the Toolbox.

I. Overview

Emissions from fires can be a significant contributor to ambient PM and ozone concentrations and visibility degradation. The contribution of primary PM emissions from smoke, such as Elemental Carbon (EC) and POA, to $PM_{2.5}$ and visibility impairment near the source is well established. However, as emissions from fires are processed in the atmosphere, secondary $PM_{2.5}$ concentrations can be formed, including ammonium sulfate and nitrate and SOA. The formation of SOA from fire emissions is complex and involves the processing of organic emissions to form SOA by many pathways (e.g., dilution and partial evaporation of the POA into gaseous species; photochemical reactions of organic species; and aerosol thermodynamics that partition the fire organics into gas-phase condensable gases and particle SOA with varying levels of volatility). The SOA component of fire $PM_{2.5}$ may not be not fully accounted for in most current modeling studies (e.g., those using the EPA National Emissions Inventory, NEI), but has been shown to be important. For example, a source apportionment analysis in Yosemite indicates fire-derived SOA during aged (2-3 days) fire plume impact periods is estimated to be ~3-4 $\mu g/m^3$ (a factor 3 to 4 times larger than the primary smoke carbon estimate (Engling et al. 2006)); A study in the Atlanta area by Lee et al. (2008) found that half of $PM_{2.5}$ organic aerosol (OA) in a smoke plume was potentially SOA representing a 1.5 to 6 times increase of the primary smoke OA contribution.

The JFSP FON Topic 3 is requesting new information and SOA_{pf} with respect to chemical species that contribute to the formations of SOA. These chemical species consist of a wide range of organic compounds with various SOA formation potentials that can change as they age. The saturation concentration (C*) is a measure of the organic compound's volatility (i.e., ability to form particle SOA) that changes as the compound oxidizes in the atmosphere. Donahue, et. al. (2009) has developed the following organic compound volatility classification:

- NVOC: Non-volatile Organic Compounds (NVOCs): These compounds reside always in the particulate phase ($C^* < 0.01$ $\mu g/m^3$).
- LVOC: Low Volatility Organic Compounds (LVOCs): These compounds are mostly in the condensed phase in all but the most remote (and warm) parts of the atmosphere ($C^* = 0.01 \sim 0.1$ $\mu g/m^3$).
- SVOC: Semi-Volatile Organic Compounds (SVOCs): Significant fractions of these compounds will be found in both phases under typical conditions ($C^* = 1 \sim 100$ $\mu g/m^3$).
- IVOC: Intermediate Volatility Organic Compounds (IVOCs): These compounds are almost entirely in the gas phase, but they comprise an enormous number of difficult to measure compounds and (probably) a small but important fraction of the total atmospheric burden ($C^* = 10^3 \sim 10^6$ $\mu g/m^3$).

[5] http://wraptools.org/
[6] http://www.wrapfets.org/
[7] http://deasco3.wraptools.org/

- VOC: Volatile Organic Compounds (VOCs): The vast majority of emissions and routinely measured organics fall in this traditional category. These are typically represented explicitly or with lumped compounds in gas-phase chemistry mechanisms ($C^* > 10^6$ µg/m^3).

The current publicly available versions of CMAQ (V5.0.2) and CAMx (V6.11) have SOA_{aqm} modules with primary organic aerosol emissions that are assumed to be non-volatile (NVOC) and gaseous VOC emissions some of which can be oxidized into gaseous Condensable Gases (CG) that are in thermodynamic equilibrium with particle SOA (CG/SOA pairs) and do not address all of the forms of SOA precursors listed above. The Volatility Basis Set (VBS) SOA_{aqm} module is a new approach for treating SOA formation that addresses the full range of partitioning and changes in volatility among organic compounds. In the past VBS has only been implemented in research grade PGMs with limited availability. More recently ENVIRON (a member of the REFERS-DMS team) has implemented VBS in the current regulatory versions of both CMAQ and CAMx (Koo et al., 2013) PGMs. Under PMDETAIL, our proposed REFERS-DMS team is developing procedures for generating fire organic EFs required for using the VBS SOA module for simulating fire SOA_{aqm} formation. Note that this requires more than just SOA_{pf} as requested by the JFSP FON, but also EFs for the whole suite of organic species (e.g., NVOC through VOC listed above as well as POA, EC, NO_X, etc.) as well as their volatility and SOA formation properties. In the current PMDETAIL project, these new VBS SOA_{pf} will be used with the new VBS version of the CAMx PGM to estimate $PM_{2.5}$ from fire emissions and the results will be compared with fire $PM_{2.5}$ estimates using the standard SOA module in CAMx. This comparison will include an evaluation against routine $PM_{2.5}$ monitoring data as well as against additional filter analysis focused on fire marker species such as levoglucosan. Routine speciated $PM_{2.5}$ measurements (e.g., IMPROVE and CSN) only measure total organic carbon (OC), don't distinguish between POA and SOA_{obs}, and it is difficult to isolate the fire organic contributions from other sources. The REFERS-DMS study will fill this void by incorporating into the Toolbox the advanced measurements from the SENEX/SEAC4RS/FRAPPE field studies, which include AMS measurements that can distinguish between POA and SOA_{obs}, that can be used to evaluate existing fire SOA_{pf}, the fire EFs developed by PMDETAIL for standard and VBS SOA_{aqm} modules, and emerging fire SOA_{pf} from new studies.

Project Benefits and Results

In the REFERS-DMS study we will provide the following benefits and results:

- Development of a summary of SOA atmospheric processes, the SOA_{aqm} modules used in current atmospheric chemistry models and the different types of fire SOA_{pf} needs that can be a resource for studies that are developing new SOA_{pf}.
- Enhancement of the PMDETAIL fire emissions modeling system to provide separate, more detailed, and more transparent treatments of emissions from fires including SOA precursors to simplify evaluation of current fire emissions models used in the PMDETAIL study, and then updating emissions based on these enhancements.
- Developing a numerical emissions and PPM/PGM modeling approach for the 2013/2014 periods during which SENEX/SEAC4RS/FRAPPE NOAA P3 field study advanced measurements have fire plume events that will form one of the bases of the Toolbox.
- Use of 2013/2014 numerical modeling laboratory and 2011 PMDETAIL modeling database (i.e., the Evaluation Toolbox) to evaluate current, PMDETAIL, and new fire SOA_{pf} using standard and VBS SOA modules and the real-world organic aerosol measurements.
- Reconciliation and refinement of modeling of the SOA_{obs} with the laboratory derived SOA_{pf} to develop new sets of SOA_{pf} that can be evaluating using the Toolbox.
- Work with EPA to transfer fire emissions modeling technology learned in REFERS-DMS to the NEI to obtain improved NEI fire emissions and allow the NEI to support emissions for the VBS SOA module.
- Collaborate with other SOA researchers funded under this FON.
- Conduct workshop/webinars with collaborators, other Smoke Science Plan participants, smoke researchers, and regulatory agencies to discuss SOA science, SOA_{pf} and results from the REFERS-DMS and other studies.
- Implement REFERS-DMS fire SOA_{pf} and EFs numerical evaluation laboratory in the WRAPTools web system.

- **Project Objectives and Hypotheses**

The REFERS-DMS proposal's tasks and research activities are aligned with JFSP FON Task 3's four objectives:

What atmospheric chemical processes transform fire emissions into SOA? Of the known chemicals emitted from fires that contribute to formation of SOA, which of their emission factors most need to be improved?

- In the first year of the study, develop a White Paper discussing atmospheric processes and species important for

fire SOA formation and the treatment of them in current atmospheric chemistry and fire emission models that can be used as a resource for REFERS-DMS and other studies to improve fire SOA_{pf} and modeling simulations.
- Apply detailed SENEX/SEAC4RS/FRAPPE measurements of fire plume impacts to evaluate fire SOA processes and species and use data analysis results to update White Paper and design fire SOA_{pf} evaluations.

To what extent do fire combustion phase, fire intensity, fuel characteristics, and type of fire (Rx or WF) influence the production of precursors to SOA?

- Enhance PMDETAIL fire emissions modeling system for the SENEX/SEAC4RS/FRAPPE study periods with fire plume events to facilitate PGM model testing of SOA_{aqm} sensitivities to combustion phase, fire intensity, fuel characteristics, and type of fire. Particular attention will be paid to the recommendation of the Smoke and Emissions Model Intercomparison Project (SEMIP) study when designing emission inventory input and output.
- Isolate fire emissions contributions to Organic Aerosol by biomass type, fire type (WF, Rx and Ag) and intensity and reconcile with SENEX/SEAC4RS/FRAPPE field study SOA_{obs} to identify potential SOA_{pf} that are most important and need more refinement.

Do the Emission factors and the chemical transformation mechanisms used in current smoke emissions and modeling systems sufficiently account for the production and aging of SOAs?

- Use emissions and chemistry modeling and detailed SENEX/SEAC4RS atmospheric measurements (i.e., the Toolbox) to evaluate whether current emissions and air quality models can simulate fire SOA processes using multiple fire SOA_{pf} and PGM SOA_{aqm} modules.
- Use results from the field experiment measurements, fire SOA_{pf} evaluation approach to identify where improvements in fire SOA_{pf} are needed.

What magnitude of change is expected in the fire National Emissions Inventory (NEI) for $PM_{2.5}$ from proposed improvements in SOA precursor emission factors?

- Fire emission inventories will be developed using current, new PMDETAIL, and emerging fire emission factors from new research for multiple representations of SOA in the atmosphere. The various versions of the 2013/2014 and 2011 fire emission inventories will allow a quantitative assessment of the effects the new EFs will have on NEI fire emissions.
- The REFERS-DMS project team will work with the EPA in implementing the improved fire SOA_{pf} and EFs in the NEI fire emissions inventory, including the ability to support the new VBS SOA_{aqm} module in CMAQ and CAMx. We have an excellent relationship with the EPA staff in charge of the NEI, successfully working with them to implement the composition and source type details of the WRAP oil and gas emissions in the 2011 NEI and to create the Graphical User Interface (GUI) and analytical tools for western states to refine/enhance their fire emissions inventories in the 2011 NEI.

Hypotheses

1) Technical methods that are in standard use in air quality planning efforts are appropriate for testing the sensitivity of PM and visibility modeling results to SOA_{pf} inputs.
2) A robust and accessible analytical and testing environment for SOA_{pf} will provide important feedback to SOA_{pf} researchers, supporting them in conducting additional research to fill important gaps in SOA_{pf} data and/or to refine SOA_{pf}.
3) The analyses done in this project and enabled by the Toolbox will support the air quality planning community by enhancing the understanding of the uncertainly of PGM SOA-related results and improving the SOA-related emission inputs and PGM modules.

II. Methods

o Study Site(s)

The Toolbox will be developed for up to four potential study sites:

2011 CONUS: The JFSP PMDETAIL study is developing a PGM modeling database for 2011 using advanced fire emissions models and the CAMx PGM. The study includes the additional analysis of filter measurements for fire markers (e.g., levoglucosan) and will identify periods with fire impacts at key $PM_{2.5}$ measurements sites in the U.S. that will be used in the REFERS-DMS evaluation Toolbox.

2013 SENEX: The SENEX field study occurred during June and July 2013 and included flights by the NOAA P3 aircraft with numerous full-science air quality measurement instruments. In particular, new measurement methods have been developed for organic aerosols (Weber et al., 2007; Jimenez et al., 2009) and their reaction intermediates

(e.g., glyoxal) (Washenfelder et al., 2011) that will allow the isolation of POA and SOA_{obs}. The SENEX NOAA P3 flights experienced several fire plume impacts that would be the focus of the REFERS-DMS fire SOA_{pf} evaluation approach. There were several other additional measurement studies operating during the SENEX field study period that will provide additional real-world data for evaluation of fire EFs in addition to the routine data from the AQS, FRM, CSN, IMPROVE and CASTNet networks:

- NOMADS[8] operated the NCAR C-130 aircraft based near Nashville during June 1 – July 15, 2013 that collected ozone, NO_X, VOC, HONO and mercury;
- SOAS[9] collected enhanced aerosol and other measurements at the Centreville, Alabama SEARCH site from June 1 through July 15, 2013.
- SEARCH[10] operates eight sites (four urban/suburban pairs) in the southeast collecting continuous gaseous and particulate measurement data for many years.

2013 SEAC4RS: The NASA Discover AQ airborne science laboratory operated a fully instrumented DC-8 aircraft that collected good data on fire plumes from the Yosemite Rim Fire as well as wildfires in Idaho during August and September 2013.

2014 FRAPPE: The FRAPPE field study occurred in July and August 2014 in the greater Denver region and included the NOAA P3 aircraft as well as the NASA Discover AQ[11] measurements (including DC8 aircraft) plus enhanced ground-based monitoring. The first FRAPPE/Discover AQ Science Team meeting is scheduled for May 2015; we will attend to assess availability of fire plume measurements from the field study.

The REFERS-DMS project team will be supplemented with an expert advisor to assist in interpretation of the airborne measurements. That individual has not been identified, but would likely be a highly-qualified, recently-retired federal employee working at a flat hourly rate.

Study Design

This study would be carried out in three phases that roughly correspond to each year of the study.

- **Year 1 / Phase I – Conceptual Design**

The first phase of the REFERS-DMS study is to prepare the conceptual design. A draft White Paper will be prepared to describe: processes in the atmosphere that affect SOA; the current and potential emerging numerical parameterizations for treating SOA in the atmosphere; what chemical species are important for SOA formation from fire emissions; and what emission factors are needed to model the formation of SOA from fire emissions. Also under Phase I we will analyze with help from an expert advisor: the SENEX, SEAC4RS, and FRAPPE field study data to identify the occurrence of fire events and develop a Modeling Protocol for developing the emissions and atmospheric chemistry numerical evaluation approach for evaluating fire SOA_{pf}. The Team will make refinements to the fire emission modeling system to more comprehensively estimate and characterize emissions from fire with particular emphasis on SOA precursors. The draft White Paper is planned to include the following elements:

- Discussion of atmospheric processes related to SOA formation.
- Description of current and potential emerging treatments of SOA_{aqm} in state-of-science atmospheric chemistry models.
- Needs for fire SOA_{pf} to support current and emerging SOA simulation modules.
- Formulation of current fire emissions models, how fire EFs are used and potential to better treat fire SOA.
- Summary of fire SOA_{pf} needs to assist researchers developing fire EFs.
- Survey of current fire SOA_{pf} and summary of researchers who are developing new fire SOA_{pf} that could be evaluated within the REFERS-DMS evaluation Toolbox.

The 2013 SENEX and SEAC4RS and 2014 FRAPPE and related field study data would be analyzed to identify periods with fire plume impacts. Particular focus will be made of fire events that are measured by the NOAA P3 aircraft advanced measurement platform. Several case studies when fire plumes are clearly evident in the observations from the 2013 and 2014 field studies will be selected, as well as from the 2011 annual PMDETAIL modeling period. A Technical Memorandum that describes the analysis of the measurement data and the selected case study periods that will be used to develop the Toolbox will be prepared. The Field Study Technical Memorandum will also describe how

[8] https://www.eol.ucar.edu/system/files/NOMADSS_OPS_PLAN_V3ag2_0.pdf
[9] http://soas2013.rutgers.edu/
[10] http://www.atmospheric-research.com/studies/SEARCH/
[12] http://www.firescience.gov/projects/08-1-6-10/project/08-1-6-10_final_report.pdf

the PGM modeling databases will be developed for the case studies and how the measurements will be used to evaluate the fire SOA_{pf}. We will survey and catalog current fire and other EFs in general and fire SOA_{pf} in particular, and post those on the REFER-DMS website along with our modeling evaluation results as they become available. An initial set of fire SOA_{pf} will be selected for evaluation in the Toolbox in collaboration with the Kreidenweis and Barsanti research teams.

The 2011 PMDETAIL fire emissions database contains information regarding fuels, moisture, combustion phase, and fire type. For this project, we will reconsider how these characteristics are combined and exported from the database to build a final inventory for input into the PGM modeling system to facilitate tagging and tracking of various fire characteristics in the model output, as well as consider the relative uncertainties of these characteristics to better inform the evaluation of SOA_{pf}. In particular, the SEMIP Phase 1 final report[12] has specific findings about fire emissions modeling uncertainties that are scale- and application-dependent; these will be considered when building the inventories for this study, as the PGM modeling will focus on specific time periods over smaller spatial scales.

We will use the enhanced fire emissions modeling system to develop new fire emissions inventories for the 2011 fire episodes in the 2011 PMDETAIL database using the most recent set of fire EFs, and for the years following 2011, to support our investigation of the episodes during the field studies which occurred in 2013/2014. The PGM model simulations will include levoglucosan as a fire marker species that would be evaluate against the special filter analysis performed as part of PMDETAIL. The fire emissions SOA_{pf} would be evaluated by comparing the modeled OA against measured values during periods of fire impacts. These preliminary results would be used to help design the remainder of the study. For the fire plume events from the 2013/2014 field study aircraft fire plume periods identified in the analysis of the field study data, we analyze meteorological observations as well as perform WRF meteorological modeling for selected events to characterize the meteorological of the fire plume events and compare WRF against the observed meteorological data.

We will update the WRAPTools website with the products from Phase I of the REFERS-DMS study, including the SOA White Paper and SENEX/SEAC4RS/FRAPPE data analysis and Toolbox design, survey and catalog of current and emerging fire EFs (including SOA_{pf}), enhanced fire modeling tools and preliminary evaluation of the current fire EFs using the PMDETAIL 2011 fire events. Collaboration is a central activity of this project and is expected to provide significant value to the JFSP. A Phase I workshop and webinar would be held to review the White Paper, discuss the results of Phase I, and refine the design for Phases II and III of the REFERS-DMS study with help from the Barsanti and Kreidenweis research groups. In addition to the project team and our collaborators, we would invite other fire researchers (e.g., EF laboratories and smog chamber researchers) and other stakeholders (e.g., air quality modelers, JFSP personnel) to the workshop/webinar where we can review SOA processes and the needs for fire SOA_{pf} and EFs in current SOA_{aqm} models.

- **Year 2 / Phase II - Build Toolbox Foundation**

Using the case studies identified from the 2013/2014 field measurement campaigns, fire emissions inventories will be built following the recommended changes and enhancements developed in Phase I. This may require obtaining and adding to the PMDETAIL database more detailed fire characteristics than were gathered for the PMDETAIL project, such as fuels, daily activity, infrared and other remote sensing data, field notes, or other information on specific fire events related to the 2013/2014 field studies. Using the initial set of fire SOA_{pf}, we would develop fire emissions for the 2013/2014 field study fire event periods and 2011 PMDETAIL fire episodes and perform a model performance evaluation. Of particular importance will be the model evaluation of the fire plumes sampled by the aircraft observations during the 2013/2014 SENEX/SEAC4RS/FRAPPE field studies. These evaluations are particularly important because: (1) the organic aerosols will be dominated by emissions from fires so we will not have the difficulty of distinguishing them from organic aerosol from biogenic and anthropogenic emissions that more dilute smoke plumes have; and (2) the aircraft measurements include AMS organic aerosol measurements that have the ability to distinguish between primary and secondary organic aerosol. ENVIRON has been successful in the past in matching the predicted and observed plume traverses using key marker species. For example, in the 1990 SOS Nashville Cumberland and the TexAQS 2006 aircraft traverses of power plant plumes, we were able to match the predicted and observed power plant plume traverses using SO_2 concentrations as a power plant plume marker species. When using TexAQS 2006 NOAA P3 measurements to estimate SO_2 oxidation rates within the Houston ship channel, we were also able to isolate the plumes using key markers species. Since fires have several species that act as good markers, we

[12] http://www.firescience.gov/projects/08-1-6-10/project/08-1-6-10_final_report.pdf

should be able to identify the periods of the aircraft traverse impacted by fire plumes using fire markers (e.g., levoglucosan, EC and OC) just like we did using SO_2 for power plant plumes. We would develop organic aerosol evaluation products for the Toolbox taking advantage of the aircraft AMS and other advanced measurements to determine the ability of the SOA_{pf} and the modeling system to reproduce the observed SOA_{obs} concentrations. The Toolbox would develop a standard set of model performance evaluation graphics that would be used to evaluate the fire SOA_{pf}. The SCICHEM PPM would be used to model all field study fire plume events and the Particulate Source Apportionment Technology (PSAT, as used in PMDETAIL) would be used to ascertain the fire SOA_{aqm} contributions by biomass type, fire intensity (flaming vs. smoldering) and fire type (WF, Rx or Ag) for selected field study events using the CAMx PGM. Information from other studies (e.g., Barsanti) that used more detailed chemical representing of fire emission processing using box modeling would be compared to and help in the interpretation of the EF evaluation using the Toolbox based on the PPM/PGM that used condensed chemical mechanisms.

We would select several initial sets of fire SOA_{pf} that would be evaluated using both the standard (CG/SOA) and VBS SOA_{aqm} module simulation approach. The fire SOA_{pf} would be evaluated using the Toolbox with the 2013/2014 field study periods and 2011 PMDETAIL fire event periods. The standard set of model evaluation products would be generated for each set of fire SOA_{pf}. At the end of Phase II we would hold another workshop and webinar with the REFERS-DMS project participants and other JFSP-funded collaborators to plan the final year of the project. More broadly, we will invite other fire researchers and go over the evaluation of the SOA_{pf} and what we have learned. We would put out a request to other JFSP researchers and other fire emission studies for fire SOA_{pf} to be evaluated using the Toolbox in Phase III.

- **Year 3/Phase III - Implement in WRAPTools**

After receipt of new sets of fire SOA_{pf} we would evaluate them using the Toolbox and post the fire SOA_{pf} and evaluation results on the WRAPTools website. We envision there might be multiple rounds of fire SOA_{pf} evaluation. The results from the evaluations would be used to identify an optimal set of fire EFs that would be evaluated again using the Toolbox. The WRAPTools website would have locations where the collaborations with the Barsanti and Kreidenweis groups, and collaborations with others from the Years 1 and 2 workshops can upload their fire SOA_{pf} (and other EFs) as well as comment on the study and download the evaluation results. A final evaluation of the most promising fire SOApf and other EFs will be evaluated using a 2014 CAMx PGM modeling database that is independent from the 2011 and 2013/2014 PGM/PPM databases used to evaluate the EFs in the evaluation Toolbox. A final workshop and webinar would be held to discuss the study and a final report and peer-review journal article prepared. In addition, we will work with EPA to transfer fire emissions modeling technology learned in REFERS-DMS to the National Emissions Inventory (NEI) to obtain improved NEI fire emissions and allow the NEI to support emissions for the VBS SOA module. We envision this could take the form of a white paper and/or direct collaboration with the EPA to incorporate the deliverables into the Emissions Inventory System (EIS).

- **Field Measurements**

No field measurements would be collected as part of REFERS-DMS study, although existing atmospheric field study measurements (SOA_{obs}) as well as fire EF (including SOA_{pf}) field measurements are integral parts of the study.

- **Data Analysis**

Akagi and others (Akagi, et al 2011) have cataloged and summarized biomass burning emission factors. Specifically, the intention of Akagi et al., 2011 was to synthesize results from a wide variety of experiments to create an ongoing summary of the best available emission factors for several basic biomass types along with typical modified combustion efficiency for those types (a surrogate for the ratio of flaming to smoldering combustion). More recently, Yokelson, et al. (2013), May et al. (2014) and Urbanksi et al. (2014) attempted to synthesize and reconcile EFs derived in the lab with those derived in the field. We propose to use the methodology and results of Akagi, May, Yokelson, Urbanski and others to create a web-based, coherent, and accessible repository for biomass burning emission factors with a focus on wildfire and prescribed burning in the United States, and particularly SOA precursors. Leveraging the technology and tools available on the WRAPTools website[13], this repository will include a portal through which others may add new research over time. In addition, we will use this repository to build an evaluation Toolbox that will allow users to evaluate and compare SOA_{pf} and EFs from different sources and export "sets" of EFs applied to emission inventories. The Toolbox will contain results from this project that apply EF sets to PGM modeling, carried through with several tags that facilitate evaluation of various fire characteristics, such as fuel type, moisture conditions, combustion phase,

[13] http://deasco3.wraptools.org

or fire type. The tools will evaluate EF performance through a series of PGM model performance analyses that use direct atmospheric measurements from multiple field campaigns, including from a "fire-centric" view that reveals performance for each of the tagged fire characteristics.

III. Project Duration and Timeline – the project would start 4 months from the date of award and last 3 years.

Project Milestone	Description	Delivery Dates
Analyze field study data	SENEX/SEAC4RS (and FRAPPE/Discover AQ when available)	6 months
White Paper	Study conceptual design describing SOA topics mentioned above.	9 months
Update WRAPTools website	WRAPTools website updated with Phase I products from REFERS-DMS study	12 months
Workshop #1	Review / discuss analysis / White Paper with researchers and users.	12 months
Update Emission Inventories	Use changes suggested in Year 1 to build new fire EIs for modeling	16 months
Modeling Simulations	Apply data in multiple modeling simulations / evaluate results.	18 months
Add results to Toolbox	Develop organic precursor emissions factors and organic aerosol evaluation products for the Toolbox	20 months
Workshop #2	Review / discuss results and analysis with researchers and users.	24 months
Add more results to Toolbox	Post, visualize and evaluate fire EFs and SOA_{pf} from REFERS-DMS Year 2 work, and from other researchers using the Toolbox	30 months
Modeling Simulations	Apply data in multiple modeling simulations / evaluate results.	32 months
Workshop #3	Review / discuss results and analysis with researchers and users.	36 months

IV. Project Compliance - NEPA and Other Clearances – Not Applicable.

V. Research Linkage

The REFERS-DMS Team recognizes and appreciates the JFSP's continued significant investment in the advancement of wildland fire and air quality science. As mentioned in the REFERS-DMS Study Design above, we plan a series of workshops with researchers and users of the information and new knowledge to be developed. We will collaborate closely with the Barsanti and Kreidenweis teams as they work on their JFSP-funded projects. The REFERS-DMS project will also leverage and coalesce the projects listed below to address the issues in Topic 3, using both already-completed studies, other studies currently underway, and other new JFSP-funded projects related to SOA precursor emissions. For the modeling, the CAMx will be fully integrated with the PMDETAIL project in progress, and the extramurally-funded 3-State Air Quality Study (3SAQS)[14] and the development of a WRAP Regional Technical Center (WRAP RTC)[15]. The 3SAQS project will assess regional air pollution transport and source contributions for Particulate Matter, Ozone, Regional Haze, and related air quality indicators in support of air quality planning by western states, using emissions, meteorological, and air quality modeling tools, source apportionment methods, and data analysis techniques. The project is currently underway, sponsored by federal agencies. The future Western RTC will complement the 3SAQS, to function on a scale similar the former WRAP Regional Modeling Center[16]. The Southeastern States Air Resource Managers have a similar regional Ozone, $PM_{2.5}$, and Regional Haze air quality analysis and planning support (SEMAP[17]) project underway. REFERS-DMS results will be shared with SEMAP to advance air quality analysis and planning in the southeastern U.S. and with the U.S. EPA Office of Air Quality Planning and Standards' (OAQPS) Modeling and Emissions Groups.

Current and pending related research grants

Grant Program	Project or Proposal Description/Identification	Funding Amount	Completion Date
JFSP Project 12-1-08-31	Synthesis of comprehensive emissions measurements and multi-scale modeling for understanding secondary organic aerosol chemistry in wildland smoke plumes	not available	September 2017
JFSP Project 14-1-03 -26	Phase Dynamics of Wildland Fire Smoke Emissions and Their Secondary Organic Aerosols	not available	August 2016

[14] http://views.cira.colostate.edu/tsdw/
[15] http://www.wrapair2.org/calendar/attachments/263/92/Western%20Regional%20Modeling%20Framework%20concept%20Nov7_2013.pptx
[16] http://pah.cert.ucr.edu/aqm/308/
[17] http://www.metro4-sesarm.org/semapabout.asp

JFSP Project 12-1-08-31	Particulate Matter Deterministic & Empirical Tagging & Assessment of Impacts on Levels (PMDETAIL)	$703,562	Fall 2015
JFSP Project 11-1-6-6	DEASCO$_3$ - Deterministic and Empirical Assessment of Smoke's Contribution to Ozone	$370,000	June 2013
JFSP Project 09-1-03-1	Experimental Determination of Secondary Organic Aerosol Production from Biomass Combustion	$621,750	December 2013
Federal / State Agencies, Private Industry	WestJumpAQMS - Modeling and Analysis of Regional Ozone and Related Air Quality Indicators	$722,000	September2013
Federal / State Agencies	3-State Air Quality Study Regional Modeling & Data Warehouse (Pilot Study 2011-14, ongoing effort 2014-present)	$1,000,000+	ongoing

VI. Deliverables and Science Delivery

- Evaluation of and recommendations on optimal SOA$_{pf}$ to apply in PGMs based on current research
- An accessible web-based portal for researchers, analysts, and others to contribute and evaluate new and existing science related to SOA$_{pf}$ development and refinement.
- Interactive web-based tools that present the results of the evaluations in the form of downloads, tables, plots, and graphics that draw connections and provide analysis on fire emissions modeling, PGM, and direct observation study results.

Always-on, web-based, secure access will be an effective and efficient way to transfer data, analytical tools, and findings of the project to fire and air quality analysts, researchers, planners, and other end users. Other REFERS-DMS deliverables include: (1) essential documentation explaining data sources, technical methods, QA/QC, results, data interpretation, and assessments of uncertainty (or other potential limitations to the technical work and findings); (2) updated fire emissions and photochemistry modules for use in PGM systems; (3) online tools and 3 workshops to orient and train potential users/reviewers of reports generated by the Toolbox; (4) presentations at technical conferences describing the Toolbox and its applications to user groups, and modeling results; and (5) a 10-20 page final report that presents the major methodologies of the project, describes significant findings, and presents analysis and recommendations.

Deliverables - description and dates

Deliverable Type	Description	Delivery Dates
White Paper and Field Studies Technical Memo	Described in text and Project Timeline above	9 months
Updated Emission Inventories		12 and 24 months
Modeling Simulations' results and evaluation		20 months and 32 months
Toolbox with SOA precursor emissions factors and organic aerosol evaluation tools /products		9 months, 24 months, 36 months
Series of 3 Workshops		12, 24, and 36 months

VII. Roles of Investigators and Associated Personnel

Roles and responsibilities of associated personnel

Personnel	Role	Responsibility
Tom Moore	Principal Investigator	Coordinate and manage project
Matthew Mavko David Randall	Technical analysis and web database support	Analysis of SOA$_{pf}$ and fire emissions, delivery of web tools, technical task oversight and science delivery
Ralph Morris Bonyoung Koo	PGM applications and technical analysis	PM and ozone modeling, evaluation, source apportionment, development of evaluation Toolbox modeling tools
Allen Robinson	Advisor on SOA$_{pf}$	Provide expert advice on evaluation of SOA$_{pf}$
To be determined	Advisor on field study data	Provide expert advice on interpreting SENEX, SEAC4RS, and FRAPPE/Discover AQ field study data
Mark Fitch, NPS	FLM collaborators	Fire activity and emissions needs of FLMs
Michael Barna, NPS		PGM applications and air quality impacts affecting protected federal lands from fire and other sources
Bret Anderson, USFS		
Bill Jackson, USFS		Interpretation of technical results for FLM use in management of air resource protection programs, as part of responsibilities for resource protection / planning
Michael George, NPS		
John Vimont, NPS		

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No.	Page	Existing Text in our Original Proposal	JFSP Reviewer Comment	Response
1	5	Technical methods that are in standard use in air quality planning efforts are appropriate for testing the sensitivity of PM and visibility modeling results to SOA_{pf} inputs. A robust and accessible analytical and testing environment for SOA_{pf} will provide important feedback to SOA_{pf} researchers, supporting them in conducting additional research to fill important gaps in SOA_{pf} data and/or to refine SOA_{pf}	Several attempts to simulating fire plumes with photochemical plume models (PGMs) reported large uncertainties due to sensitivity to and uncertainty of wind speeds and wind directions. When modeled winds (e.g., WRF winds) are compared to local wind measurements the differences are typically significant. The resolution of PGMs and their plume parameterizations are not adequate for fire plumes. These uncertainties would confound the evaluation of the emission factors and SOA modules.	For developing the EF evaluation toolbox using the aircraft traverse fire plume impacts from the field studies, we have added the use of the SCICHEM photochemical puff model (PPM) that includes many of the same chemical mechanisms as the PGMs, but has more flexibility in defining its transport and dispersion characteristics. We will use fire markers (e.g., levoglucosan) to define the locations of the fire impacts along the aircraft measurement traverse and then define the winds and dispersion inputs to SCICHEM to match the observed fire plume extent. SCICHEM can then be used to cost-effectively analyze multiple SOApf definitions and its chemistry will photochemically process the fire SOApf emissions to determine the PM2.5 and SOA impacts along the observed aircraft traverse. SCICHEM will be applied for all fire aircraft impacts and the CAMx PGM will also be setup for some of the most promising fire plume aircraft measurement traverses for the further evaluation of the alternative fire EFs.
2	N/A		SOA formation in a fire plume is a much more complex problem than SO_2 oxidation in a power plant plume with well known emissions and stack exit parameters. While SO_2 chemistry is well documented, there are still wide gaps in our understanding of fire plume dynamics and SOA chemistry.	The discussion of the example of how we have evaluated power plant plume SO4 formation rates using aircraft measurements was not meant to imply that SOA formation is in any way comparable to SO4 formation as SOA formation is a much more complicated process. Rather, it was to demonstrate that we have experience in using source markers (SO_2 in this case) to define plume impacts along aircraft measurement traverses so that photochemical models can be used to evaluate photochemical processing in the plumes. Similar plume isolation techniques along the aircraft measurement traverse can be used for fire plumes using fire markers.
3	N/A		They will explore aqueous phase chemistry in the models, but there are still significant uncertainties here and it is not clear a modeling study will reduce this.	To the extent that clouds are present in the 2011 and 2013/2014 modeling periods, then aqueous-phase chemistry modules will be invoked in the photochemical models in the toolbox. Aqueous-phase SOA chemistry is less well understood and we would document the extent it is present in

			our toolbox modeling databases.
4	N/A	Will exercise and evaluate existing CTMs. However, there are already known limitations and deficiencies in these modeling systems. The new field data available to evaluate the models will allow the evaluation of intermediate products in the CTMs organic chemistry mechanism. However, the causes of deviation will likely not be known due to the lumped mechanism used in the models. Consequently, while this proposed project is valuable and there is a need to better understand current model performance, box modeling studies with detailed representations of the physical-chemical processes, as opposed to the lumped processes in current CTMs, may result in more useful information for further development of emission inventories and modeling of total organic aerosols from biomass burning emissions.	There is a trade-off between detailed modeling of the chemistry of many species and simplifying other atmospheric processes, versus using a lumped chemical mechanism but having a more complete representation of the other atmospheric processes (e.g., background chemistry conditions, three-dimensional transport and dispersion, deposition, etc.). We find using both techniques yields the best results and we will collaborate with Kelley Barsanti's group who are using a box model to analyze fire SOA EFs.
5	N/A	The PIs are offering to assemble existing EF information with photochemical models that are commonly used to build an SOA testing and evaluation toolbox. The concept is very attractive, but the PIs would depend on existing measurements or those that may come out in the literature, and these are insufficient to solve the questions underlying the issue the FON was crafted to address. Until the fire sciences community can deliver more refined EFs for SOA precursors, and model formation mechanisms that can reproduce observations with those EFs, the toolbox will be of limited use. This is an effort that should certainly be undertaken, however, once better data and mechanisms become available.	Several new fire SOA EF development studies have been funded by the JFSP starting in 2014. We will collaborate with these studies (e.g., Barsanti and Kreidenweis) and evaluate these new SOA EFs using the evaluation toolbox and catalog them within the WRAPTools EF library.