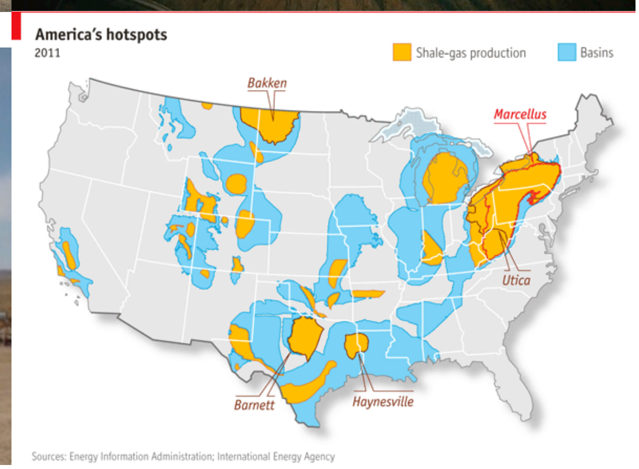
Shale Oil and Natural Gas Nexus (SONGNEX)

Studying the Atmospheric Effects of Changing Energy Use in the U.S. at the Nexus of Air Quality and Climate Change



A NOAA Field Study in the Western U.S. in April-May of 2015

Introduction

Energy production and use in the U.S. have seen rapid changes over the past decade. The domestic production of oil and natural gas has grown strongly, natural gas is increasingly replacing coal for the generation of electrical power, and the contribution from renewables has rapidly grown (Fig. 1). Many of these shifts have caused significant changes in the atmospheric emissions of trace gases and fine particles that are at the root of the Nation's air quality and climate change issues. However, since the changes in emissions are poorly known, the net effects for air quality and climate change are still very uncertain. Over the past decades, the U.S. has effectively addressed air quality issues and it is important to assure that the changes in our energy infrastructure do not negate some of these positive changes. Likewise, as the Nation is increasingly focused on mitigating the effects of climate change, it is important to know the net changes in emissions of greenhouse gases and other trace gases and fine particles that force the climate to change.

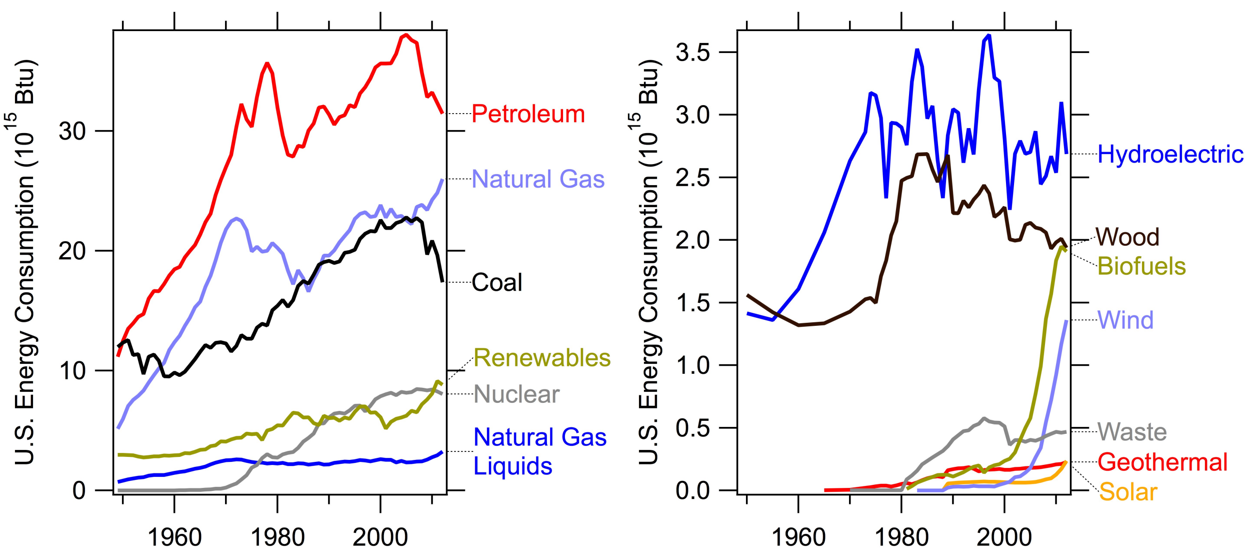


Figure 1: Energy consumption in the U.S.: all sources on the left and renewable energy sources on the right (data from the Energy Information Administration). Energy production is very similar, except for petroleum, of which a large fraction is imported

The domestic production of oil and natural gas from tight sand and shale formations is one change in our energy system that has received particular attention recently. Technological advances in directional drilling and hydraulic fracturing have made the production of oil and natural gas from tight sand and shale formations economically viable, and the production has increased strongly over the last decade (Fig. 2). This increased production of oil and natural gas has been associated with emissions of methane, non-methane hydrocarbons and other trace gases to the atmosphere. The concerns about these emissions are three-fold:

- **Climate:** Increased emissions of methane, a potent greenhouse gas, and black carbon aerosol could lead to a net increase in the radiative forcing of climate.

- **Air quality:** Emissions of methane, non-methane hydrocarbons, and nitrogen oxides can lead to the formation of ozone and fine particles in the atmosphere, which deteriorate air quality.
- **Air toxics:** Emissions of air toxics could have direct health effects.

These three issues will be discussed in more detail below.

In the SONGNEX mission described here, we will quantify the atmospheric emissions from various components of the U.S. energy infrastructure. We will also study how these emissions transform chemically in the atmosphere, and how these transformations contribute to ozone and fine particle formation. Specifically, we intend to focus on the emissions from a number of different tight oil and shale gas basins in the western U.S. The basins are chosen to represent a significant fraction of the production as well as different stages of the development of a basin. In addition, we will study several other components of the energy infrastructure, including surface coalmines in Wyoming, coal and natural gas power plants, a major intersection of crude oil pipelines in Oklahoma, and biofuel refineries where ethanol is made from corn.

The results of SONGNEX will give the information that is needed to (1) evaluate the effects of various energy sources and uses on climate, air quality and human health, and to (2) allow communities and society as a whole to make the best decisions to minimize these effects.

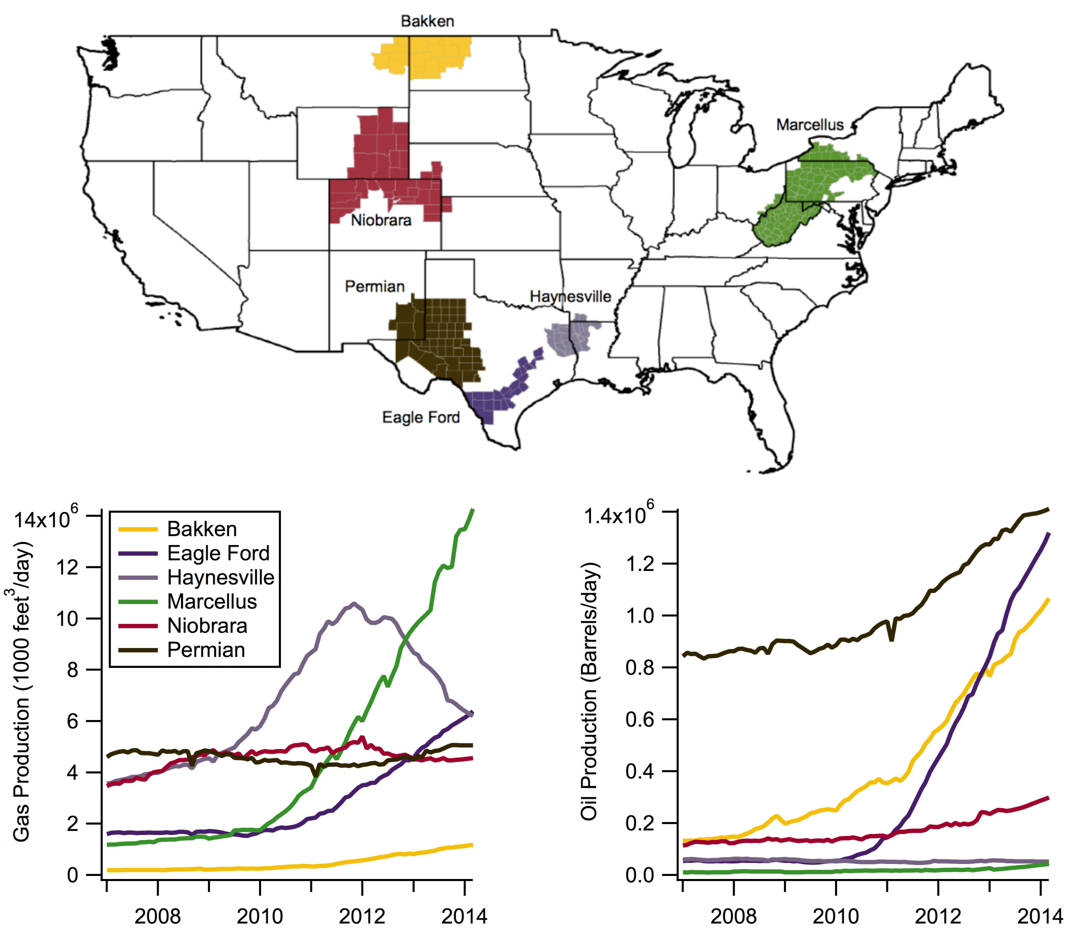


Figure 2: Oil and natural gas production in a few key tight oil and shale gas regions (Energy Information Administration).

Background: Atmospheric Effects of Shale Oil and Natural Gas

Climate

A significant fraction of natural gas consists of methane and the release of some of this potent greenhouse gas during the different stages of natural gas production and use is common [*Brandt et al*., 2014]. Several studies have compared natural gas to coal as en energy source, and pointed out that the net greenhouse gas emissions from natural gas can be higher than from coal if the methane leaks are too high [*Howarth et al*., 2011; *Wigley*, 2011; *Alvarez et al*., 2012]. These analyses are not without controversy [*Cathles et al*., 2012; *Howarth et al*., 2012], but part of the discussion is caused by the fact that the net emissions of methane during the different stages of shale oil and natural gas production are still very poorly quantified and understood. Two studies led by the NOAA Global Monitoring Division put the methane leak rate at 2.3-7.7% of production in the Denver-Julesburg basin in Colorado [*Petron et al*., 2012], and at 6.2-11.7% in the Uintah basin in Utah [*Karion et al*., 2013]. A study led by the University of Texas put the methane leak rate from several types of sources much lower at 0.42% [*Allen et al*., 2013]. At present, it is unknown to what extent these differences are caused by differences between these basins in raw gas composition, industry practices, regulation, or the methods employed in the different scientific studies, i.e. the two NOAA estimates were top-down as they used ambient methane to estimate the total emissions, whereas the University of Texas study was bottom-up as it estimated the total emissions from the sum of known sources. To answer these questions, it will be essential to extend measurements of methane emissions to other basins with consistent measurement and analysis approaches, and also to do such studies at different stages of basin development.

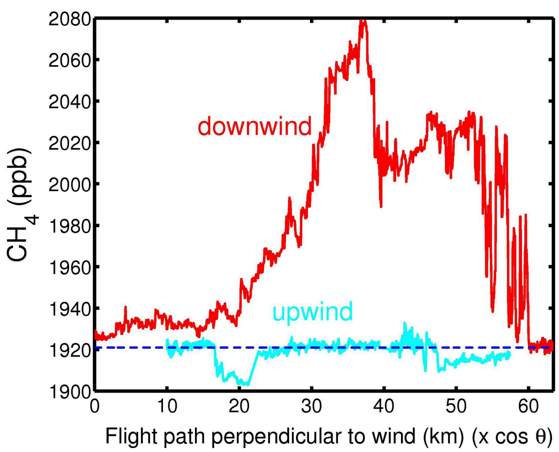


Figure 3: Airborne measurement of methane up- and downwind from oil and gas production fields in the Uintah basin in Utah [*Karion et al*., 2013]. Methane fluxes were determined from the difference in integrated methane between the up- and downwind plumes, multiplied by the orthogonal wind speed and depth of the atmospheric boundary layer.

Emissions of black carbon from natural gas flaring provide a second climate issue related to oil and natural gas production. A recent assessment concluded that black carbon aerosol constitutes the second most important human emission in terms of its climate forcing in the present-day atmosphere [*Bond et al*., 2013]. However, the emissions of black carbon from natural gas flaring are highly uncertain. One inventory estimated BC emissions from flaring at nearly 4% of the anthropogenic global total with the majority originating in Russia, Nigeria, and the Middle East [*Bond et al*., 2013]. Another study quantified the emission of black carbon aerosol from natural gas flares and estimated that the total amounts to 1.6% of global BC emissions from energy related combustion [*McEwen and Johnson*, 2012].

Air Quality

The release of methane, non-methane hydrocarbons, nitrogen oxides and several other trace gases from oil and natural gas production can contribute to the formation of ozone and fine particles in the atmosphere, two air pollutants that also exert a radiative forcing of the climate system. An early study demonstrated the enhanced concentrations of hydrocarbons in an oil and gas production region in Texas and Oklahoma [*Katzenstein et al*., 2003]. A recent analysis for the Marcellus region suggested an enhancement in nitrogen oxides [*Carlton et al*., 2014]. The effect of these emissions on ozone formation was most vividly demonstrated in the Upper Green River basin in Wyoming [*Schnell et al*., 2009; *Carter and Seinfeld*, 2012] and the Uintah basin in Utah [*Edwards et al*., 2013], where exceptionally high levels of ozone can be formed in the winter, when strong inversions trap the emissions near the surface during cold-pool conditions (Fig. 4). In the Colorado Front Range, an area that has recently gone into non-attainment for ozone, it was shown that the emissions from oil and gas production can contribute more than half of the hydrocarbon reactivity, suggesting that these emissions could be important for summer ozone formation [*Gilman et al*., 2013; *Swarthout et al*., 2013]. A modeling study for the Haynesville shale region in Texas and Louisiana showed that the 8-hour ozone design value could increase by as much as 5 ppbv due to the emissions associated with natural gas production in the region [*Kemball-Cook et al*., 2010]. This would make it much more difficult for the area to meet the National Ambient Air Quality Standard (NAAQS).

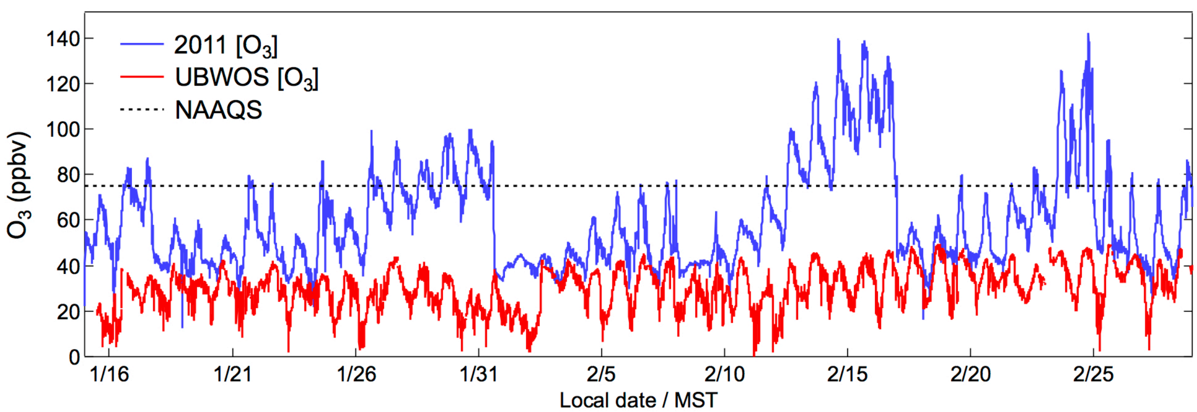


Figure 4: Winter ozone formation in the Uintah basin in 2011 (blue) compared to ozone in 2012 when no cold-pool conditions were present in the basin [*Edwards et al*., 2013]. The dotted line represents the National Ambient Air Quality Standard (NAAQS), which was exceeded on multiple days in 2011.

Air Toxics

Significant concerns regarding the emissions from oil and natural gas production are related to the release of air toxics such as hydrogen sulfide (H_2S) and the so-called BTEX compounds (benzene, toluene, ethyl benzene and xylenes). In the Colorado Front Range, it was shown that oil and gas production could be responsible for a significant fraction of ambient benzene [*Petron et al*., 2012; *Gilman et al*., 2013], and a study in Garfield County, Colorado quantified the human health risks due to such emissions [*McKenzie et al*., 2012].

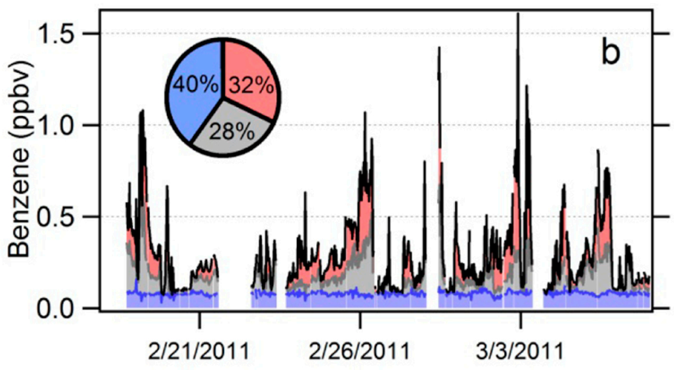


Figure 5: Measured time series of benzene at the Boulder Atmospheric Observatory, with the estimated contribution from oil and gas production in red and the contribution from vehicular emissions in grey [*Gilman et al.*, 2013]. The contribution in blue reflects the background that is transported from elsewhere.

NOAA Study in the Western U.S. in Spring 2015

Tight oil and shale gas basins

The primary goal of the NOAA SONGNEX study proposed here is to quantify the emissions of trace gases and fine particles from several different tight oil and shale gas basins in the western U.S., and to study the chemical transformation of these emissions. The study will be focused on the following basins, which represent a mixture of oil and gas production regions at various stages of development.

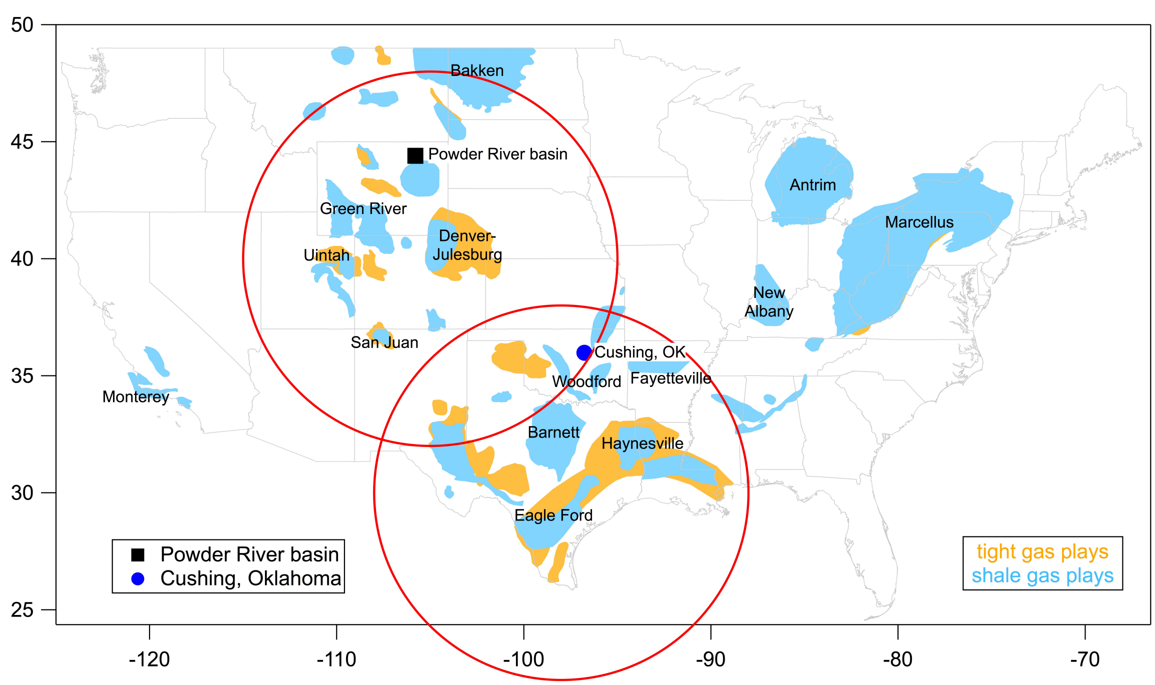


Figure 6: SONGNEX flight objectives. Tight sand and shale plays are shaded. The black square shows the location of surface coalmines in the Powder River Basin, Wyoming. The blue circle shows the location of a major intersection in crude oil pipelines near Cushing, Oklahoma. The open red circles show the range of the NOAA WP-3D for flights that take off and land from airfields in the Denver area in Colorado and a location to be determined in Texas.

- **Bakken, North Dakota:** Oil production is growing rapidly in this region (Fig. 2). Due to a relative lack of pipeline infrastructure, a significant fraction of the crude oil is transported by rail, which has led to significant accidents. Also, the light from gas flares can be observed from space [*Elvidge et al.*, 2009] and recent observations have revealed a strong increase in flaring in the Bakken (Fig. 7).
- **Upper Green River Valley, Wyoming:** This is the basin where winter ozone formation was first observed [*Schnell et al.*, 2009].
- **Uintah basin, Utah:** Winter ozone formation was also observed in this basin and the subject of three intensive Uintah Basin Winter Ozone Studies (UBWOS 2012, 2013 and 2014) led by the NOAA Chemical Sciences Division [*Edwards et al.*, 2013]. Methane emissions from the basin were estimated to be high at 6.2-11.7% of production [*Karion et al.*, 2013].
- **Denver-Julesburg basin, Colorado:** Multiple studies in the Colorado Front Range by NOAA GMD and CSD scientists and others have investigated the emissions of methane, hydrocarbons and air toxics in the area [*Petron et al.*, 2012; *Gilman et al.*, 2013; *Swarthout et al.*, 2013]. Natural gas production in this area has been relatively stable (Fig. 2), but can be expected to increase in 2014.
- **Haynesville shale, Texas:** Natural gas production in this area has declined in recent years due to a reduction in drilling activity. Measurements of emissions were estimated from two research flights with the NOAA WP-3D during the SENEX study in summer 2013, and appeared to be lower than reported for Utah and Colorado as a fraction of production (unpublished results). These findings need to be confirmed, and continued measurements may give insight into the relation between emissions and drilling activity.
- **Eagle Ford, Texas:** Oil production is growing very rapidly in this region (Fig. 2). As in the Bakken, gas flares are common and can be observed from space (Fig. 7).
- **Marcellus, Pennsylvania:** Natural gas production is rapidly growing in this basin and it is now the largest producer of shale gas in the U.S. (Fig. 2). Methane emissions were estimated from one flight with the NOAA WP-3D during SENEX in 2013, and appeared to be the lowest among the other basins studied (unpublished results). Time permitting, we will consider additional flights in the region.

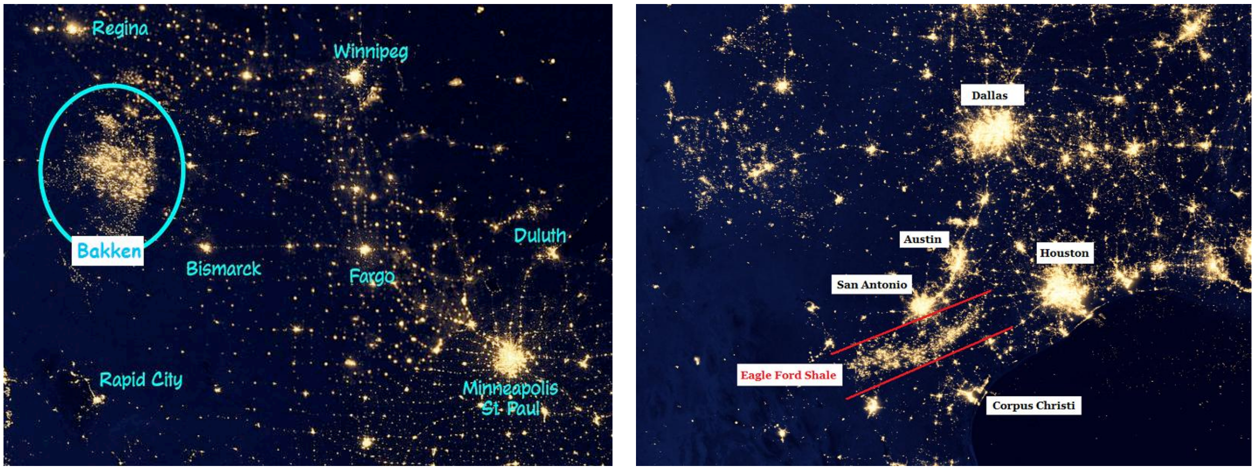


Figure 7: Gas flaring in the Bakken in North Dakota (left) and Eagle Ford shale in Texas (right) as observed from the Visible Infrared Imaging Radiometer Suite (VIIRS) satellite instrument (NOAA National Geophysical data Center)

Other parts of the U.S. energy system

Various other components of the U.S. energy infrastructure will also be targeted during SONGNEX:

Surface coalmines: Mining in the Powder River basin in Wyoming provided about 45% of coal in the U.S. in 2012 (Fig. 8). Most of this coal is transported to other states by rail. Atmospheric emissions from surface mines are more difficult to quantify than from underground mines. An earlier study found that the methane emissions are proportional to the length of the exposed seams [*Kirchgessner et al*., 2000]. In view of the intense discussion regarding net greenhouse gas emissions from coal vs. natural gas [*Howarth et al*., 2011], it is of interest to revisit these measurements by making them from aircraft. In addition, it is important to quantify the emissions of other trace gases and fine particles.

- **Crude oil pipelines:** The increased domestic production of crude oil requires additional pipeline capacity. One of the major crossroads in the U.S. pipeline system for crude oil is at Cushing, Oklahoma (Fig. 9). We propose to measure emissions from this complex during SONGNEX.
- **Coal and natural gas power plants:** Power plants in the U.S. are increasingly using natural gas instead of coal, and emissions of CO_2, nitrogen oxides and sulfur dioxide have significantly decreased as a result [*de Gouw et al*., 2014]. Building on previous work on power plant emissions, we will compare a few coal and natural gas power plants in the western U.S. in terms of emissions as well as ozone and PM formation in the plumes.
- **Biofuel refineries:** At present, roughly 10% of the volume of gasoline sold in the U.S. consists of ethanol made from corn. This ethanol is produced in over 200 biofuel refineries. Most of these are in the mid-West, but there are also several plants in Colorado whose emissions will be investigated as part of SONGNEX.

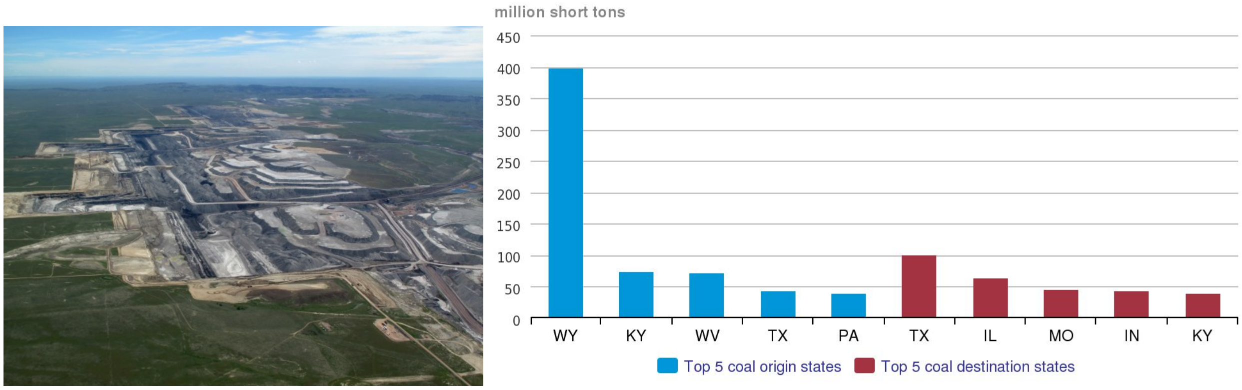


Figure 8: Surface coalmines in the Powder River Basin, Wyoming (left) and the top 5 coal origin and destination states in 2012 (right) (Energy Information Administration).

Figure 9: Crude oil pipelines in North America (left) (Energy Information Administration). Tank batteries at Cushing, Oklahoma, one of the major intersections in the pipeline system (right).

Opportunities for additional research

Other emission sources that may be studied include biomass burning: the western U.S. has experienced several years with multiple large wildfires. These fires affect air quality and potentially climate through the scattering and absorption of sunlight by the fine particles that are emitted and formed downwind from the fire. While the SONGNEX study will not be during the seasonal maximum of the fire season, there have been early fires in previous years. Should a wildfire occur in the Rocky Mountain States, the NOAA WP-3D will undertake a flight to sample the emissions and study the plume chemistry. These results would provide a preliminary data set for a larger biomass burning study that CSD scientists plan to propose beyond 2015.

Collaborations and synergies

We will seek out collaborations with several other groups within and external to NOAA that have the following expertise:

- **Mobile laboratories:** Several groups including the NOAA Global Monitoring Division have the ability to field measurements in mobile laboratories that contain a subset of the measurements on the NOAA WP-3D. Simultaneous measurements from a mobile laboratory within the basin and from an aircraft flying over the basin provide the most detailed insight into emission sources.

- **Network measurements:** Several groups including the NOAA Global Monitoring Division as well as States and regulatory agencies field regular network measurements in some of the basins of interest. The long-term nature of these measurements provide a powerful data set to address how changes in drilling activity and oil and gas production have affected emissions and ambient concentrations of hydrocarbons.
- **Oil sands and natural gas research in Canada:** We have ongoing collaborations with the group of Dr. Shao-Meng Li at Environment Canada, which is tasked with the environmental research on the Alberta oil sands [*McLinden et al*., 2012], as well as tight oil and natural gas production in Canada. Dr. Shao-Meng Li spent a sabbatical at NOAA in 2013, and was a collaborator in the NOAA Uintah Basin Winter Ozone Studies in 2013 and 2014. Initial discussions have started regarding collaboration with Environment Canada in the framework of SONGNEX.
- **Satellite measurements:** Several groups are researching the use of methane measurements from satellite instruments [*Parker et al*., 2011; *Schneising et al*., 2011] to quantify the fluxes associated with oil and gas production. In addition, methanol is used by the industry as an anti-freeze in pipelines, and is released in large quantities in Utah. Methanol can be measured from satellites [*Wells et al*., 2012] and is another direction to explore in collaboration with colleagues.
- **Modeling:** Several groups have begun to model the effects of oil and gas production on regional air quality. We will make our data available to those groups and work closely together with them on the interpretation of the data.
- **States:** We have worked with several states (e.g. Utah, Colorado, Texas) on their air quality issues and will continue these collaborations in the framework of SONGNEX to assure that the information is widely available to the stakeholders.
- **Industry:** We have done part of our previous work on winter ozone formation in collaboration with industry groups, which has been mutually beneficial. We will continue to communicate with these groups regarding mutual research interests.

SONGNEX Science Questions

By performing flights over the areas described in the previous section, we plan to answer the following detailed science questions:

1. Climate: What is the effect of shale oil and natural gas production on emissions of radiative forcing agents?

- What are the emissions of methane from different shale basins?
- What are the methane emissions as a fraction of the basin production and relative to the drilling activity?

- What is the relative importance of distributed sources at wellheads versus centralized processing plants? What other sources of methane are present (agricultural, landfills)?
- What determines the methane emissions from shale oil and gas production? What are the relative roles of raw oil and gas composition, industry practices, drilling activity and regulation?
- Can we detect black carbon aerosol from gas flaring in the Bakken and Eagle Ford? If so, what are the emission strengths and is gas flaring a significant source of black carbon aerosol?

Approach: For methane fluxes, box patterns around the shale regions of interest will be flown to integrate the horizontal fluxes going into and coming out of the box (see Fig. 3). Correlation with other species (hydrocarbons) will be used to distinguish between natural gas and sources. For black carbon, exploratory flight legs near the areas where the density of flaring is the highest will be conducted. If significant emissions are found, box patterns to quantify them will be flown.

2. Air quality: What are the emissions of ozone and aerosol precursors and how are they chemically transformed in the springtime atmosphere?

- What are the emissions of hydrocarbons from different shale basins?
- How do the hydrocarbon emissions compare between different basins, and how do they relate to raw oil and gas composition, industry practices and regulation?
- What is the relative importance of distributed sources at wellheads (e.g. condensate tanks, dehydrators) versus centralized processing plants?
- What are the emissions of nitrogen oxides? What is the relative importance of combustion sources at wellheads (separators, generators, pump jacks) versus mobile sources?
- What are the chemical transformations of these emissions in the springtime atmosphere and how do they contribute to ozone and fine particle formation?

Approach: The same box patterns as for the methane fluxes will address these questions. Additional downwind flight legs will be used to further study chemical transformations.

3. Air Toxics: What are the mixing ratios of BTEX compounds and hydrogen sulfide?

- What are mixing ratios and emissions of BTEX compounds (benzene, toluene, ethyl benzene, xylenes) in different shale basins?
- What fraction of ambient BTEX compounds can be attributed to oil and natural gas production?
- What are the mixing ratios of hydrogen sulfide in different shale basins?
- What is the relative importance of sources at wellheads versus evaporation ponds (where those are used)?

Approach: Flight legs over shale basins will be used to determine the distribution in mixing ratios of different air toxics. High time-resolution measurements will be used to identify specific sources.

4. Other questions: What are the emissions from other components of the U.S. energy infrastructure?

- What are the emissions associated with surface coal mining? How do these emissions compare to emissions from natural gas production?
- What are the emissions from coal and natural gas power plants? How do the chemical transformations in the plumes compare?
- What are the emissions associated with the transport of crude oil by pipeline, and specifically from the major crossroads in the pipelines?
- What are the emissions from biofuel refineries? How do these emissions compare with other stages of the lifecycle of a biofuel?

Approach: Flight legs downwind from these sources will allow quantification of these emissions.

The NOAA WP-3D Research Aircraft and its Proposed Payload

We propose to use the NOAA WP-3D research aircraft to answer the science questions discussed above. The ability of the aircraft (i) to reach a number of different shale regions within the same study, and (ii) to carry a large enough instrument payload to address all of the science questions is unique and cannot be provided by any other platform.

We propose to operate the NOAA WP-3D out of Rocky Mountain Metropolitan airport in Broomfield, Colorado for part of the mission, as well as out of an airport in Texas. To complete some of the more remote flight objectives (e.g. Bakken, Marcellus), the NOAA WP-3D may have to end that day's mission close to the area of interest, refuel, and return the next day while targeting additional science objectives en route. Towards that goal we will investigate one additional airfield in North Dakota and one in Pennsylvania, and the potential to park and access the NOAA WP-3D and add consumables for the scientific instruments.

The proposed payload for the SONGNEX mission is described in the table below. Final choices will depend on instrument availability, investigator interest, and funding.

Parameter	Method
Gas Phase	
Carbon dioxide (CO_2); methane (CH_4)	Infrared laser absorption
$^{13}CH_4$	Cavity ring-down spectroscopy
Carbon monoxide (CO)	VUV fluorescence
Nitrogen oxides (NO, NO_2, NO_y)	O_3 chemiluminescence
Ozone (O_3)	NO chemiluminescence
NO, NO_2, NOy, O_3	Cavity ring-down
Hydrogen sulfide (H_2S)	Cavity ring-down spectroscopy
Sulfur dioxide (SO_2)	Pulsed UV fluorescence
Ethane (C_2H_6)	Quantum cascade laser absorption
Non-methane hydrocarbons	Whole air sampler (WAS) & post-flight GC-MS analysis

Hydrocarbons and oxygenates	Proton-transfer time-of-flight mass spectrometry (PTR-ToF)
Formaldehyde (HCHO)	Laser-induced fluorescence
Glyoxal (CHOCHO)	Cavity-enhanced absorption spectroscopy
PANs	I^- CIMS with heated inlet
Nitric and nitrous acid (HNO_3, HONO)	I^- CIMS
Ammonia	Acetone dimer CIMS
Aerosol Phase	
Size-resolved non-refractory aerosol composition	Compact time-of-flight aerosol mass spectrometry (AMS)
Single particle black carbon	Single particle soot photometer (SP2)
Aerosol number, surface and volume distribution	Ultra high sensitivity aerosol spectrometer (UHSAS), condensation particle counters (CPC), optical particle counters (OPC)
Other	
Meteorological parameters	Various sensors

Table 1: Proposed payload of the NOAA WP-3D research aircraft during SONGNEX.

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