



Project 039 Naphthalene Removal Assessment

Massachusetts Institute of Technology

Project Lead Investigator

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University Participants

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- PI(s): Prof. Steven R. H. Barrett and Dr. Raymond Speth (Co-PI)
- FAA Award Number: 13-C-AJFE-MIT, Amendment Nos. 026, 034, 043, and 053
- Period of Performance: July 8, 2016 to August 31, 2020 (with the exception of funding and cost share information, this report covers the period from October 1, 2018 to September 30, 2019)
- Task(s):
 1. Explore relationship between polycyclic aromatic hydrocarbon (PAH) formation and aircraft particulate matter (PM) emissions
 2. Calculate air quality and climate impacts of naphthalene removal
 3. Conduct integrated cost-benefit analysis of impacts of naphthalene removal in the United States

Project Funding Level

The funding comprises \$840,000 in FAA funding and \$840,000 in matching funds. Sources of match are approximately \$233,000 from Massachusetts Institute of Technology (MIT), plus third-party in-kind contributions of \$361,000 from Oliver Wyman Group and \$246,000 from Byogy Renewables, Inc.

Investigation Team

- Prof. Steven Barrett (MIT) serves as PI for the A39 project as head of the Laboratory for Aviation and the Environment. Prof. Barrett both coordinates internal research efforts and maintains communication among investigators in the various MIT research teams mentioned below.
- Dr. Raymond Speth (MIT) serves as co-PI for the A39 project. Dr. Speth directly advises students performing research in the Laboratory for Aviation and the Environment, with a focus on assessment of naphthalene removal refinery options; climate and air quality modeling; and fuel alteration life-cycle analysis. Dr. Speth also coordinates communication with FAA counterparts.
- Prof. William Green (MIT) serves as a co-investigator for the A39 project, as head of the Green Research Group. Prof. Green advises students on work in the Green Research Group focused on computer-aided chemical kinetic modeling of PAH formation.
- Mr. Randall Field (MIT) is the Executive Director of the MIT Energy Initiative and a co-investigator of the A39 project. Drawing upon his experiences as a business consulting director at Aspen Technology Inc., Mr. Randall provides mentorship to student researchers in the selection and assessment of naphthalene removal refining options and process engineering at large.
- Mr. Drew Weibel (MIT) is a graduate student researcher in the Laboratory for Aviation and the Environment. Mr. Weibel is responsible for conducting selection and assessment of naphthalene removal refining options; calculation of refinery process requirements and fuel composition effects from selected processes; estimation of



capital and operating costs of naphthalene removal processes; air quality and climate modeling; and integrated cost-benefit analysis.

- Mr. Lukas Brink (MIT) is a graduate student researcher in the Laboratory for Aviation and the Environment. Mr. Brink is responsible for the development of a combustor model quantifying the effect of naphthalene removal on soot emissions, the modeling of air quality and climate impacts, and integrated cost-benefit analysis.
- Mr. Max Liu (MIT) is a Ph.D. candidate in the Green Research Group. Mr. Liu is responsible for development and analysis of a chemical kinetic model of PAH formation with fuel-composition effects.
- Dr. Mica Smith (MIT) is a postdoctoral associate in the Green Research Group. Ms. Smith is responsible for the experimental measurements being used for the validation of the chemical kinetic mechanisms.
- Dr. Agnes Jocher (MIT) is a postdoctoral associate in the Green Research Group. Ms. Jocher is responsible for evaluating microphysical models that link the presence of PAH molecules to the formation of soot particles and for providing modeling expertise in combining these models with the kinetic models being developed.

Project Overview

Aircraft emissions impact the environment by perturbing the climate and reducing air quality, thus leading to adverse health impacts, including increased risk of premature mortality. As a result, understanding how different fuel components can influence pollutant emissions, as well as the resulting impacts and damage to human health and the environment, is important in guiding future research aims and policy. Recent emissions measurements have shown that removal of naphthalenes while keeping the total aromatic content unchanged can dramatically decrease emissions of particulate matter (Brem et al., 2015; Moore et al., 2015). The objective of this research is to determine the benefits, costs, and feasibility of removing naphthalenes from jet fuel, with regard to the refiner, the public, air quality, and the environment. Specific goals of this research include:

- Assessment and selection of candidate refining processes for the removal of naphthalenes from conventional jet fuel, including details of required technology, steady-state public cost, and changing life-cycle emissions impacts at the refinery
- Development of a chemical kinetics model to better understand the link between fuel aromatic composition and the resulting PM emissions due to jet fuel combustion
- Assessment of the climate and air quality impacts associated with naphthalene reduction and/or removal from jet fuel
- Development of a life-cycle analysis of the relative costs of removing naphthalene from jet fuel and the associated benefits due to avoided premature mortalities and climate damage for a range of possible scenarios

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Task 1 - Explore Relationship Between PAH Formation and Aircraft PM Emissions

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Objective

The formation of black carbon (soot) from hydrocarbon fuels can be considered to take place in two stages. First, fuel components and combustion intermediates react and form PAHs. Large PAHs then act as soot nuclei, which grow as they absorb both PAH and other species, coagulate through collisions with other soot particles, carbonize, and partially oxidize (Richter & Howard, 2000). The details of the fuel composition mainly affect the first step of this process: the formation of PAHs. The objective of this task is to develop a combustor model that includes the formation of PAH species from different fuel components and the conversion of these PAH species to soot particles or non-volatile PM (nvPM) emissions, to enable evaluation of the sensitivity of soot emissions to fuel composition.



Research Approach

The Reaction Mechanism Generator (RMG) was used to develop a detailed chemical kinetic mechanism for jet fuel combustion that includes the formation of PAH (Gao et al., 2016). As part of this task, we have extended RMG to include recently discovered PAH growth pathways, such as the phenyl addition pathway, cyclization via carbenes, and an aromatic-catalyzed intramolecular H-transfer mechanism. We then used this improved version of RMG to generate a higher-fidelity chemical kinetic model for the formation of PAHs in naphthalene-containing flames. This approach is being utilized to produce a chemical kinetic mechanism describing the first stage of soot particle production.

These mechanisms are then utilized within Cantera (Goodwin et al., 2013), a combustion modeling framework. A combustor model, as schematically shown in Figure 1, is developed to evaluate emissions for varying fuel compositions and flight conditions. The model consists of two main parts: the primary zone and the secondary zone. In the primary zone, upstream air is mixed with fuel and reacts with a certain characteristic residence time. Then, in the secondary zone, the resulting air-fuel mixture is mixed with both secondary and dilution air.

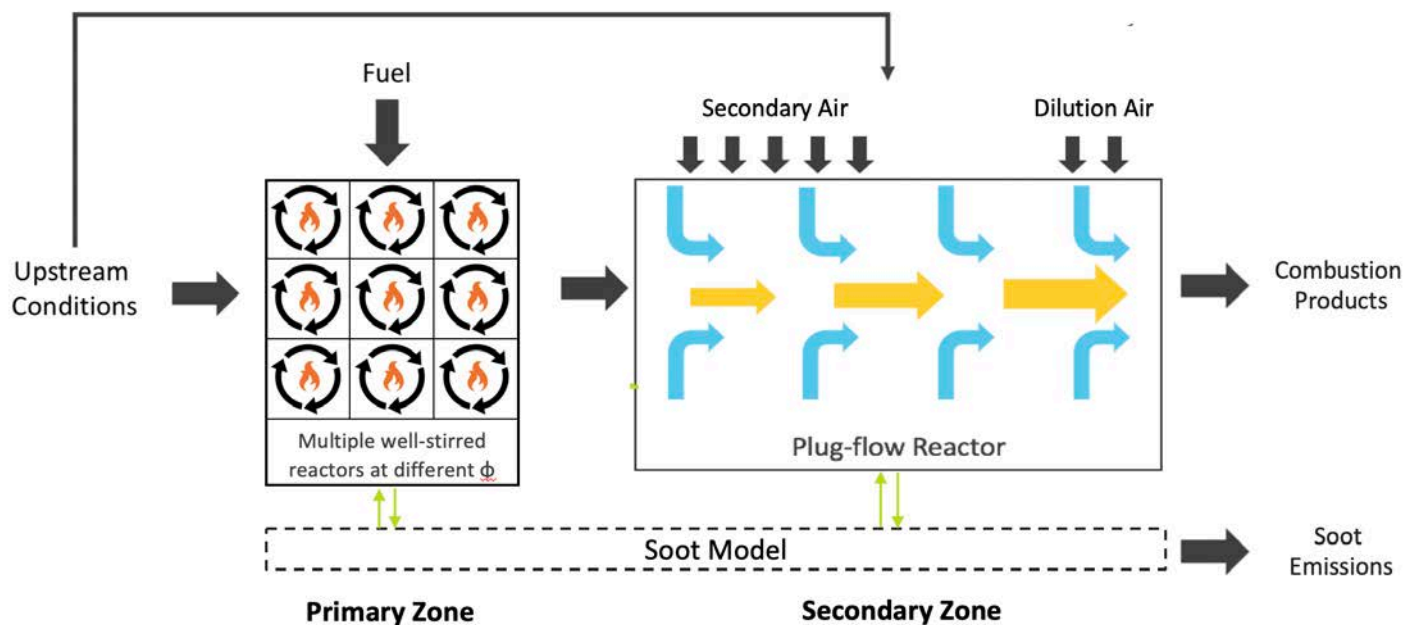


Figure 1. Schematic of the gas turbine combustor model including a soot model.

The primary zone consists of a set of N well-stirred reactors at different equivalence ratios, to account for mixture inhomogeneity. The secondary zone is modeled as a plug-flow reactor and is shown in more detail in Figure 2. After the flow exits the N primary zone reactors, it is mixed again and split into an outer and core part upon entering the secondary zone. This latter split is used to model the incomplete mixing that occurs in the second part of the combustor. The fraction of the air-fuel mixture that enters the secondary-zone core is a model variable, as are the location along the secondary zone where both streams become fully mixed and the pattern in which the mixing takes place. Other design variables in the secondary zone include the division between secondary and dilution air, the starting location of both, and the length over which they are injected.

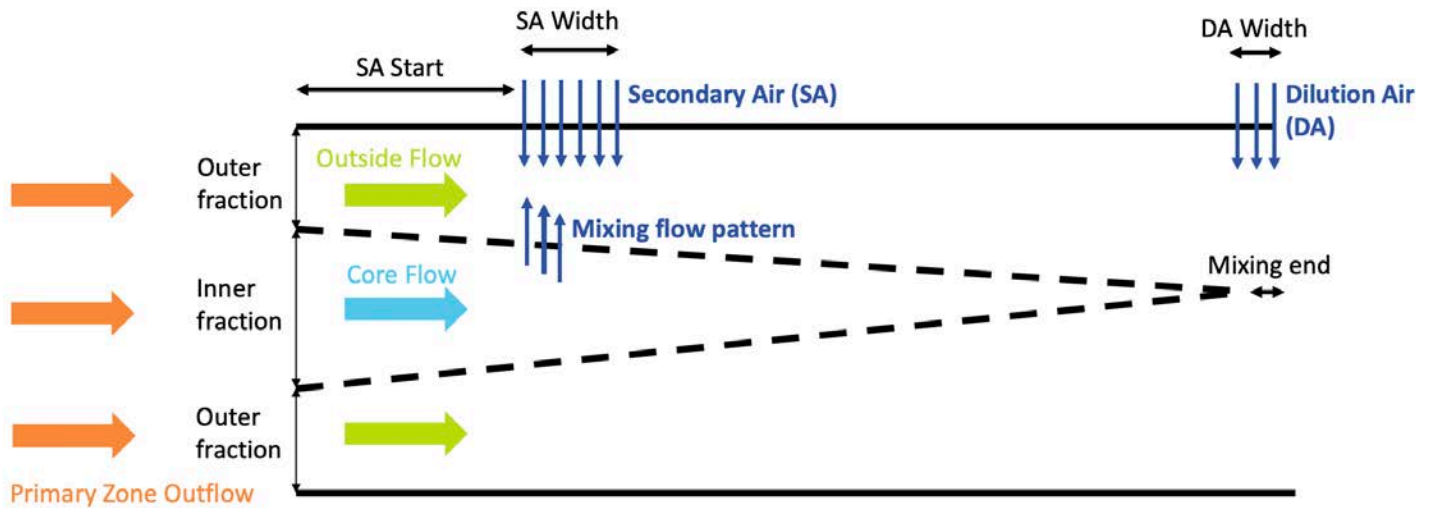


Figure 2. Overview of the combustor model secondary zone.

As shown in Figure 1, the primary and secondary zones are both coupled with a soot model. This soot model consists of four stages: nucleation (*nuc*), coagulation (*coag*), surface growth (*sg*), and oxidation (*ox*). For each of the four stages, multiple mechanisms from the literature are implemented, and the corresponding effects on the soot number (*N*) and mass (*M*) density are computed. Using multiple mechanisms provides confidence that the obtained results are robust even though the individual mechanisms from the literature may vary. This is especially important, because all mechanisms are simplifications of a complex problem, and caution is thus warranted when they are used outside the specific cases and conditions for which they were originally developed and tested.

Equation 1 shows the dependency of the number density on the four stages of the soot formation process. Because surface growth does not affect the total number of particles, no surface growth term is present.

$$\frac{dN}{dt} = \left(\frac{dN}{dt}\right)_{nuc} + \left(\frac{dN}{dt}\right)_{coag} + \left(\frac{dN}{dt}\right)_{ox} \quad (1)$$

The mass density development is shown in Equation 2. No coagulation term is present here because coagulation through collisions of soot particles is not expected to decrease the overall soot mass.

$$\frac{dM}{dt} = \left(\frac{dM}{dt}\right)_{nuc} + \left(\frac{dM}{dt}\right)_{sg} + \left(\frac{dM}{dt}\right)_{ox} \quad (2)$$

The implemented nucleation mechanisms can be divided into three categories, depending on the main precursor species used. The most extensive methods first model the dimer formation from PAH species and then use the dimer concentration to calculate the nucleation rates. The second set of methods directly considers the PAH species and their collision rates. Third, a set of methods is implemented by using the C_2H_2 concentration, which is assumed to be directly correlated with PAH concentrations (Schiener & Lindstedt, 2018).

For coagulation, two main mechanisms are considered. The first, from Kazakov and Frenklach (1998), splits coagulation into a pure coagulation part and an aggregation part, and considers collision frequencies in the free, transition, and continuum regimes. In the more straightforward second method, the coagulation rate is computed on the basis of a single collision frequency rather than multiple frequencies in different regimes (Wen et al., 2003).

Similarly to coagulation, the surface growth mechanisms can also be divided into two categories. The first category consists of methods modeling surface growth as being a first-order reaction with acetylene (C_2H_2) and being proportional to the total soot surface area. The second category combines surface growth and oxidation by using the Hydrogen Abstraction C_2H_2 Addition (HACA) mechanism (Frenklach & Wang, 1991). This mechanism considers six main reactions and uses them to compute the active radical carbon site number density, which then determines the soot surface growth and oxidation rates.

Beyond the HACA mechanism oxidation method, a second group of implemented oxidation methods directly considers the concentrations of O_2 , OH , and O as well as the total soot surface area in computing the change in soot mass density. In order to calculate the effect of oxidation on the soot number density, a log-normal particle size distribution (Zhao et al., 2003) is assumed, relating the decrease in soot mass density to a decrease in soot number density.

The formation of soot adds extra complexity to the overall combustor model, because in addition to the previously existing gas phase, an additional solid (soot) phase is present. Because Cantera solely covers the former, the interactions between both phases must be accounted for manually. This process is shown in more detail in Figure 3. During nucleation and surface growth, carbon and hydrogen from the gas phase to the solid phase in the form of PAH species and C_2H_2 . However, oxidation causes carbon and hydrogen to shift back from the solid phase to the gas phase in the form of CO_2 , CO , and H .

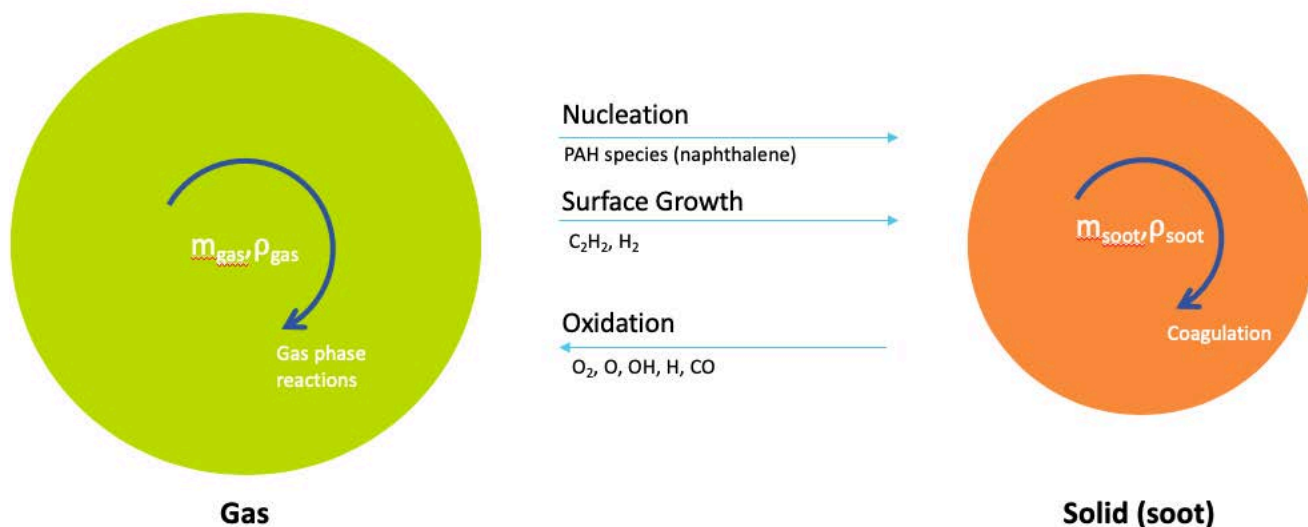


Figure 3. Interactions between the gas phase and the solid (soot) phase in the combustor model.

Emission data from the ICAO Aircraft Engine Emissions Databank (EASA, 2019) are used to optimize the primary and secondary zone design parameters to accurately match NO_x and CO emissions for take-off, climb, idle, and approach flight conditions. Sweeps using different combinations of soot mechanisms as well as fuel compositions are performed next to generate data on the sensitivity of soot emissions to varying naphthalene concentrations.

Milestone(s)

Both the primary and secondary zone of the combustor model were completed, and the different soot mechanisms were implemented.

Major Accomplishments

The chemical kinetic mechanism and combustor model were used to calculate formation rates of PAH species over a range of engine-relevant conditions. Figure 4 shows the evolution of several key aromatic species as the engine thrust setting is varied from near-idle to full power conditions for a representative surrogate fuel.

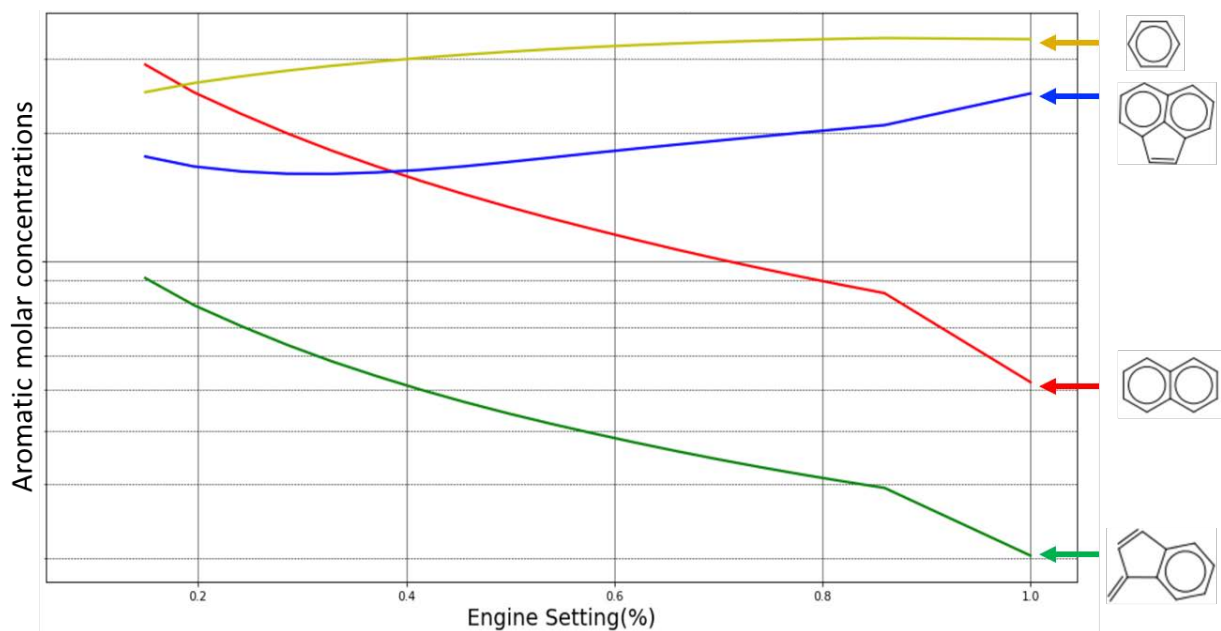


Figure 4. Evolution of selected aromatic species concentrations in the combustor primary zone as a function of engine setting.

Publications

N/A

Outreach Efforts

N/A

Awards

None.

Student Involvement

This task was conducted primarily by Drew Weibel and Lukas Brink, working directly with Prof. Steven Barrett and Dr. Raymond Speth. Mr. Weibel graduated with a Master of Science in 2018.

Plans for Next Period

The next step for this task is to optimize the primary and secondary zone design parameters to match emission data. After this optimization is performed, naphthalene concentrations in the fuel can be varied, and data from the various soot mechanisms on the effects of different fuel compositions on soot emissions will be collected.

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Task 2 - Calculate Air Quality and Climate Impacts of Naphthalene Removal

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Objective

The objective of this task is to calculate the air quality and climate impacts of a policy in which naphthalene is removed from jet fuel used in the United States.

Research Approach

The air quality effects of changes in aircraft PM emissions are evaluated by using the GEOS-Chem adjoint model, which we have previously used for assessing health impacts of emissions (Dedoussi & Barrett, 2014). The use of an adjoint model, which is a computationally efficient approach to calculating the sensitivity of an aggregate objective function (e.g., population exposure to PM_{2.5}), enables evaluation of a range of scenarios in a single run, thus allowing for incorporation of upstream uncertainty in the emissions indices for different species. The PM exposure calculated by using GEOS-Chem includes both the effects of changes in black carbon emissions and changes due to sulfur reductions that accompany the removal of naphthalenes (in the case in which hydrotreating is used to remove naphthalenes). The spatial pattern of emissions of nvPM, and sulfur compounds is taken from the 2015 inventory from the Aviation Environmental Design Tool (AEDT).

Climate impacts of naphthalene removal include contributions at both the fuel production and fuel consumption stages.

The additional refinery processing required to reduce or remove naphthalene requires process fuel, steam, electricity, and, in the case of hydrotreating, hydrogen production. The greenhouse gas (GHG) emissions associated with each of these processes increase life-cycle jet fuel GHG emissions. Using the results calculated as part of the refinery modeling work conducted in the previous project year, we found the GHG emissions associated with naphthalene removal to be 135 g CO₂e per kg fuel for hydrotreating and 144 g CO₂e per kg fuel for extractive distillation.

Consumption of reduced-naphthalene fuel decreases radiative forcing (RF) from aviation black carbon, and reductions in sulfur decrease the cooling effect of sulfates (Mahashabde et al., 2011). Contrail impacts are estimated according to studies on the impact of reducing the number of ice nuclei available for contrail formation. Caiazza et al. (2017) have found that decreasing ice nuclei by 67% (an amount representative of a fully paraffinic biofuel) reduces contrail RF by <13%. Burkhardt et al. (2018) found that reducing ice nuclei by 50% reduces contrail RF by ~20%. Here, the reductions in contrail RF found in these studies are scaled by the estimated reduction in nvPM emissions from naphthalene removal.

The combined climate impacts of these effects are evaluated by using the APMT-Impacts Climate model, a policy-oriented rapid assessment tool that provides probabilistic estimates of climate impacts.

Milestone

The work completed for this task was presented in an Information Paper prepared for the CAEP/12-WG3/2 meeting and presented on October 7, 2019.



Major Accomplishments

On the basis of a literature review of nvPM emissions measurements from engines using fuels with varying levels of naphthalene (Brem et al., 2015; DeWitt et al., 2008), the potential range of reduction in nvPM emissions associated with 95% naphthalene removal was estimated to be 15–40%, or 5.0–12.5 mg nvPM per kg fuel. Monetized climate impacts for the different climate forcing pathways are summarized in Table 1, presented on a cents-per-gallon basis with both median values and a range indicating the 90% confidence interval. Monetized air quality impacts of naphthalene removal are similarly summarized in Table 2.

Table 1. Monetized climate benefits of naphthalene removal.

Impact Pathway	Impact (¢/gallon)
Black carbon radiative forcing (15% nvPM reduction)	0.09 (90% CI: 0.01 – 0.23)
Black carbon radiative forcing (40% nvPM reduction)	0.23 (90% CI: 0.04 – 0.61)
Contrail radiative forcing (15% nvPM reduction)	1.06 (90% CI: 0.30 – 2.59)
Contrail radiative forcing (40% nvPM reduction)	2.77 (90% CI: 0.77 – 6.89)
Hydrotreating CO ₂ emissions	-1.82 (90% CI: -0.30 – -4.70)
Extractive distillation CO ₂ emissions	-1.89 (90% CI: -0.31 – -5.01)
Sulfate aerosol (hydrotreating only)	-4.17 (90% CI: -0.61 – -11.23)

Table 2. Monetized air quality benefits of naphthalene removal.

Impact Pathway	Impact (¢/gallon)
nvPM emissions (15% nvPM reduction)	0.04 (90% CI: 0.02 – 0.06)
nvPM emissions (40% nvPM reduction)	0.11 (90% CI: 0.06 – 0.16)
Sulfur emissions (hydrotreating only)	1.92 (90% CI: 1.04 – 2.76)

Publications

N/A

Outreach Efforts

N/A

Awards

None.

Student Involvement

This task was conducted primarily by Drew Weibel, working directly with Prof. Steven Barrett and Dr. Raymond Speth.

Plans for Next Period

Additional work has been planned to estimate changes in contrail radiative forcing associated with the use of naphthalene-depleted fuels by using the Contrail Evolution and Radiation Model (Caiazzo et al., 2017). In addition, current air quality results are based on a regional atmospheric model, simulated at a resolution of 0.5° × 0.667°, whereas nvPM impacts have been shown to be underestimated with lower-resolution models (Punger & West, 2013). Additional work has been planned to provide better estimates of these impacts through higher-resolution local modeling approaches.

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Task 3 - Conduct Integrated Cost-Benefit Analysis of Impacts of Naphthalene Removal in the United States

Massachusetts Institute of Technology

Objective

The objective of this task is to produce an integrated cost-benefit analysis of naphthalene removal in the United States, accounting for the additional refining cost as well as the air quality and climate impacts.

Research Approach

The overall cost-benefit assessment of naphthalene removal includes fuel production costs, air quality benefits, and climate impacts from fuel production and fuel consumption. These effects are placed on a common monetized basis to compare different naphthalene removal scenarios. We consider uncertainties in the assessment of each component and use these uncertainties to compute the likelihood of a net benefit for different scenarios.

Milestone

The work completed for this task was presented in an Information Paper prepared for the CAEP/12-WG3/2 meeting and presented on October 7, 2019.

Major Accomplishments

The processing costs, air quality benefits, and climate impacts of naphthalene removal are converted to a common basis of cents per gallon, as presented in Table 3. The totals shown exclude contrail effects, which have not yet been quantified. In the absence of large impacts on contrail net radiative forcing, the current results suggest that the benefits of widespread naphthalene removal are outweighed by the costs of processing the fuel and the CO₂ emissions associated with that processing.



Table 3. Costs (positive) and benefits (negative) of naphthalene removal.

	Component	Hydrotreatment (¢/gallon)		Extractive Distillation (¢/gallon)	
Processing	Refinery	9.1	(8.7 – 9.5)	6.4	(6.1 – 6.8)
Air quality	nvPM	-0.1	(-0.02 – -0.16)	-0.1	(-0.02 – -0.16)
	Fuel sulfur	-1.9	(-1.0 – -2.7)	0	
Climate	nvPM	-0.2	(-0.02 – -0.6)	-0.2	(-0.02 – -0.6)
	Fuel sulfur	3.9	(0.6 – 10.5)	0	
	Contrails	-1.7	(-0.4 – -5.3)	-1.7	(-0.4 – -5.3)
	Refinery CO ₂	1.8	(0.3 – 4.5)	1.8	(0.3 – 4.8)
Total		10.9	(5.8 – 16.8)	6.3	(2.4 – 9.3)

For both naphthalene removal processes, the climate impacts of the refinery CO₂ emissions exceed the air quality and climate benefits of naphthalene removal, neglecting the potential for significant contrail effects. In addition, in the case of hydrotreatment, the net present value of the climate warming associated with sulfur removal is greater than the NPV of the reduced air-quality-related damages. In addition to these environmental costs are the costs associated with processing jet fuel in the refinery. These results suggest that, in the absence of a strong contrail effect, naphthalene removal on a nationwide basis would unlikely to be cost beneficial. However, naphthalene removal may still be beneficial under certain circumstances, e.g., if applied to fuels used at airports with particular air quality concerns.

Publications

N/A

Outreach Efforts

The results of this work were presented to CAEP Working Group 3 at a meeting in Garching, Germany on October 7, 2019.

Awards

None.

Student Involvement

This task was conducted primarily by Drew Weibel, working directly with Prof. Steven Barrett and Dr. Raymond Speth. Mr. Weibel graduated with a Master of Science in 2018.

Plans for Next Period

Future work for this task consists of incorporating updated climate and air quality impacts, as well as evaluating scenarios in which naphthalene is removed only at certain places or times to maximize the benefit, e.g., targeting fuel used at specific airports.