Quantification of Aldehydes Emissions from Alternative and Renewable Aviation Fuels using a Gas Turbine Engine

Hu Lia\*, Mohamed A. Altahera, Chris W.Wilsonb, Simon Blakeyb, Winson Chungb, Lucas Ryeb

aEnergy Research Institute, Leeds University, Leeds, UK

bDepartment of Mechanical Engineering, Sheffield University, Sheffield, UK

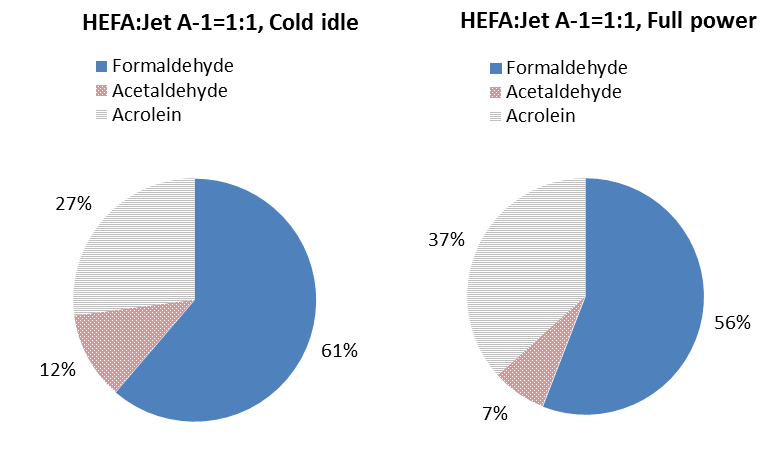
## Corresponding Author

Dr Hu Li, Energy Research Institute, School of Process, Environmental and Materials Engineering, Leeds University, Leeds, LS2 9JT, UK. Email: [fuehli@leeds.ac.uk](mailto:fuehli@leeds.ac.uk). Tel: 0044 113 3437754. Fax: 0044 113 2384.

# ABSTRACT

In this research three renewable aviation fuel blends including two HEFA (Hydrotreated Ester and Fatty Acid) blends and one FAE (Fatty Acids Ethyl Ester) blend with conventional Jet A-1 along with a GTL (Gas To Liquid) fuel have been tested for their aldehydes emissions on a small gas turbine engine. Three strong ozone formation precursors: formaldehyde, acetaldehyde and acrolein were measured in the exhaust at different operational modes and compared to neat Jet A-1. The aim is to assess the impact of renewable and alternative aviation fuels on aldehydes emissions from aircraft gas turbine engines so as to provide informed knowledge for the future deployment of new fuels in aviation. The results show that formaldehyde was a major aldehyde species emitted with a fraction of around 60% of total measured aldehydes emissions for all fuels. Acrolein was the second major emitted aldehyde species with a fraction of ~30%. Acetaldehyde emissions were very low for all the fuels and below the detention limit of the instrument. The formaldehyde emissions at cold idle were up to two to threefold higher than that at full power. The fractions of formaldehyde were 6~10% and 20% of total hydrocarbon emissions in ppm at idle and full power respectively and doubled on a g/kg-fuel basis.

**Keywords:** aldehyde emission; formaldehyde emission; gas turbine; renewable aviation fuel; alternative aviation fuel.



# INTRODUCTION

Air transport has been steadily increasing by ~5% per year globally over the past three decades and forecasted to continue to increase in the next decade ([Belobaba et al., 2009](#_ENREF_2)). With the increase in demand for air transport, emissions from aircraft engines have to be monitored and controlled to protect public health and the environment, particularly in the vicinity of airports.

There are a variety of air pollutants emitted from aircraft engines that can potentially affect human health and the environment. Common species such as NOx (Nitrogen Oxides), CO (Carbon Monoxide), UHC (Unburned Hydrocarbon) and smoke are regulated by ICAO (International Civil Aviation Organization) ([ICAO, 2008](#_ENREF_11)) and considered as primary species in emission inventories. CO2 as a greenhouse gas is considered as a global concern rather than a local issue. There is an increasing concern and need to include additional emission species which have potential health and environmental concerns in emission inventories. These species are often called HAPs (Hazardous Air Pollutants), which are part of VOCs (Volatile Organic Compounds). ICAO reported some examples of HAPs that have been identified as representative pollutants from airport sources including formaldehyde, acetaldehyde, acrolein, 1,3-butadiene, benzene, Naphthalene, toluene, xylene and propionaldehyde ([ICAO, 2011](#_ENREF_12)). These compounds play an important role in the atmospheric chemistry and urban air quality ([ICAO, 2011](#_ENREF_12), [Leikauf, 2002](#_ENREF_17), [Koenig, 2000](#_ENREF_15)) and have major health concerns. They are also precursors to free radical ozone productions ([Lea-Langton et al., 2009](#_ENREF_16)). The research for aviation activity related or sourced HAPs is at early stages and knowledge on these HAPs emissions is very limited ([ICAO, 2011](#_ENREF_12)). It is reported that formaldehyde, acetaldehyde, acrolein and methyl ethyl ketone are the main species of carbonyl emissions from the engine exhaust and are toxic, mutagenic and even carcinogenic to human body ([He et al., 2009](#_ENREF_9), [Pang et al., 2006](#_ENREF_23)). Both formaldehyde and acetaldehyde were classified as a probable human carcinogen by EPA in 1987 and exposure to them can cause irritation of the eyes, nose and respiratory tract ([EPA, 1989](#_ENREF_7)). Formaldehyde has a strong potential to form ozone and is classified as an active ozone formation precursor. Ozone is irritant gas that can pose hazards to mucous membranes of eyes and respiratory tract.

Knighton et al ([Knighton et al., 2007b](#_ENREF_14)) measured 20+ VOCs including aldehydes and benzene using the tunable infrared laser differential absorption spectroscopy for formaldehyde and the proton transfer reaction mass spectrometer for other VOCs from CMF56-2-C1 engines of a DC-8 aircraft during APEX program using three different fuels: two batches of JP8 containing 17.5% and 21.8% aromatics respectively and a high sulfur jet fuel. They found that formaldehyde and benzene concentrations were hardly detectable at the higher engine thrust conditions, until the engine thrust was reduced to 15% or lower, where they started to increase quickly as the engine thrust continued to reduce to idle. Li et al ([Li et al., 2011](#_ENREF_18)) assessed aldehydes emissions under atmospheric pressure and 600K using a radial swirler industrial low NOx gas turbine combustor and compared aldehydes emissions between B100 (100% Waste cooking oil Methyl Ester or WME), B20 (80% Kerosene: 20% WME) and pure kerosene. In their experiment, the same FTIR (Fourier Transform Infrared) instrument as in this paper was used to determine aldehydes including formaldehyde, acetaldehyde and acrolein. Their results showed that formaldehyde was the most prevalent aldehyde species for all the fuels, accounted for up to 50%.

There has been an increasing interest in the development of alternative fuels for aviation due to energy supply security concerns and potential environmental benefits (CO2 reduction). The alternative aviation fuels include both synthetic and renewable jet fuels ([D. Bulzan et al., 2010](#_ENREF_6)). Synthetic fuels, also called FT-SPKs (FT- Synthetic Paraffinic Kerosenes), are derived from coal (CTL-Coal To Liquid) or natural gas (GTL-Gas To Liquid) via the FT (Fisher Tropsch) process. Extensive researches and tests have been done on these fuels for the certification purposes. The blends of up to 50/50 (by volume) of FT-SPK with JP-8 or Jet A have been approved in the USA for the US military and civil aviation use ([MIL-DTL-83133H, 2011](#_ENREF_21)). However, these alternative fuels are not renewable as coal and natural gas are fossil fuels. Although biomass can be used in the FT process to produce BTL to make it renewable, the cost is high and the availability of large amount of biomass is a question ([Rye et al., 2010](#_ENREF_24)). The interests therefore have been moved to Hydrotreated Renewable Jet (HRJ) or Hydrotreated Vegetable Oil (HVO). HRJ or HVO is hydrocarbon aviation fuels produced from vegetable/plant oils or animal fats via hydroprocessing. This type of fuel is also called bio-SPK, “green jet” or HEFA (Hydrotreated Ester and Fatty Acid).

All alternative/renewable aviation fuels are required to be drop-in fuels ([Blakey et al., 2009](#_ENREF_3)), which mean that they must be completely interchangeable and compatible with conventional kerosene based jet fuels and can be added to conventional jet fuels as a substitute without a need to modify the engine and fuel system. These alternative/renewable aviation fuels have no or trivial amount of aromatic hydrocarbons and sulphur ([Corporan et al., 2011](#_ENREF_5)), which are an advantage over conventional kerosene based fuels because of its benefits on the reduction of particulate matter emissions ([Lobo et al., 2011](#_ENREF_19)). Lobo et al ([Lobo et al., 2012](#_ENREF_20)) compared regulated gaseous and particulate matter emissions from the same engine as in this paper between CTL, GTL fuels and Jet A-1 and reported a significant reduction in PM emissions by CTL and GTL fuels and remarkable hydrocarbon reductions by GTL fuel. Christie et al ([Christie et al., 2012](#_ENREF_4)) measured PAH emissions from the same engine using neat CTL, neat GTL fuels and 50/50 GTL/Jet A-1 blend and reported a significant reduction by GTL but an increase by CTL. There are other potential benefits from these alternative fuels that need to be identified. This forms the objective of this paper, i.e. to assess and quantify aldehydes emissions of three renewable aviation fuel blends and one alternative fuel (GTL), and compare them with conventional Jet A-1 fuel.

# EXPERIMENTAL

## Fuels

The neat conventional kerosene based Jet A-1 was used as the reference fuel. Three renewable fuel blends and a neat GTL fuel (can be used as a component of aviation fuel) were tested. Table 1 shows the blending ratio and selected chemical and physical properties of each fuel. Two separate fuel tanks were used with one for Jet A-1 and the other for testing fuels.

**Table 1.** Selected properties of the fuels tested

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Fuel code | Fuel blending ratio | Specific heat  (MJ kg-1) | Sulphur  (ppm) | H/C Ratio | Density  (kg m-3) | Aromatics  (wt%) |
| Jet A-1 | Jet A-1  100% | 43.2 | 669 | 1.89 | 803.5 | 16 |
| E | 100% GTL | 44.4 | <5 | 2.19 | 737.9 | ~0 |
| F | HEFAa:Jet A-1  50:50 | 43.5 | 335 | 2 | 780 | 8 |
| H | FAEb:Jet A-1  10:90 | 42.18 | 601 | 2 | 810 | 15.5 |
| I | HEFA:Jet A-1  75:25 | 43.9 | 181 | 2.1 | 763.6 | 4.5 |

a:HEFA-Hydrotreated Ester and Fatty Acid

b: FAE: Fatty Acids Ethyl Ester

## 2.2 Engine

An Artouste MK113 APU (Auxiliary Power Unit) engine was used as a test bed for the emission measurements. It is a single spool gas turbine engine, in which a centrifugal compressor is driven by two stage turbine through a single rotating shaft. All operating parameters of the engine such as fuel flow rate, RPM, exhaust temperatures and pressure and fuel consumption were monitored and recorded throughout the tests. Table 2 presents the selected nominal operational values for the APU at idle and full power for all the fuels.

**Table 2.** APU engine operating conditions

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Power mode | RPM | Fuel flow rate  (kg h-1) | EGTc  ⁰C | Air fuel ratio |
| Idle  Full power | 20200~20800  34200~34400 | 51~53  115~116 | 500~506  520~546 | 65~67  68~70 |

c:EGT-Exhaust Gas Temperature

## 2.3 Emission measurement

## 2.3.1 Overall set up

A stainless steel plate was fixed behind the APU exhaust so that the installed sampling probes would sit no further than half an exhaust diameter behind the engine exhaust plane as required per the Aerospace Recommended Practice (ARP) ([SAE-Aerospace, 2004](#_ENREF_25)). Two sample probes were mounted on the steel plate, one for gaseous emission measurement and the other for PM measurement (not presented in this paper). The exhaust gas samples for gaseous measurements were transported to instruments via a heated sample transfer line maintained at 150⁰C to avoid water vapor condensation and to minimize heavy hydrocarbon condensation. A schematic of the setup for the study is presented in figure 1 excluding particulate matter measurement.

During the test, the APU was started and stabilized using Jet A-1 for few minutes and then switched to testing fuels. The engine was set to run approximately 6 minutes at idle (cold idle) and 6 minutes at full power and then returning to idle (hot idle). The operating parameters of the engine were monitored and recorded (table 2).

APU

FID, NDIR, Chemiluminescence

FRF FTIR

THC

NOx

CO/CO2

FTIR

Hydrocarbon speciation

Figure 1 Schematic view of overall sampling system setup

## 2.3.2 Gaseous emission measurements and aldehydes speciation

Gaseous emissions were measured by a FID (Flame Ionization Detector) for THC (Total Hydrocarbon), a NDIR (Non-Dispersive Infrared) for CO and CO2, and a chemiluminescence for NOx. All these instruments were calibrated using calibration gas bottles (zero and span gases) each time when a new fuel was to be tested.

A Gasmet CR-2000 Fourier Transform Infrared (FTIR) was used to detect and quantify three aldehydes: formaldehyde, acetaldehyde and acrolein, along with 35 other calibrated VOC compounds. The FTIR is capable of measuring concentrations as low as 0.5-3 ppm depending on species and applications with 2% accuracy of the measurement range. The calibration ranges for formaldehyde, acetaldehyde and acrolein were 0~100, 0~200 and 0~500 ppm respectively. The exhaust sample was drawn at the same position as that for regulated gaseous emission analyzers by a heated sample line with its temperature being controlled at 180⁰C. The sample flow rate was 2-3 l.min-1. The samples were analyzed at a frequency of 0.5Hz. At least two hundred measurements for each testing condition were taken and the average values were reported. One of the main objectives of using the FTIR in this study is to get specified hydrocarbon information, including alkenes, aromatic hydrocarbon, and aldehydes. This paper is focused on aldehydes emissions due to their strong ozone formation potentials.

## 2.3.3 Emission index calculation

The emission measurements were on a volumetric basis. These were converted into emission index using equation 1 ([SAE-Aerospace, 2004](#_ENREF_25), [Li et al., 2011](#_ENREF_18)):

As and ; thus:

g/kg-fuel Equation 1

Where; mi is the mass rate of the emission component i; Ci is the concentration of the component; Di is the density of an emission component i; Ve is the volumetric flow rate of the exhaust gas; me is the mass flow rate of the exhaust gas; De is the density of the exhaust gas (air density is used here); AFR is the air/fuel ratio on a mass basis calculated by carbon balance; mf and ma are the fuel and air mass flow rate respectively; MWi and MWa are the molecular weight of the emission component i and air respectively.

# RESULTS and DISCUSSIONS

## 3.1 Aldehydes emissions as a function of engine power and warm up

Aldehydes emissions are the products of incomplete combustion and are related to combustion temperatures and thus engine power settings. The EI (Emission Index) of three aldehydes: formaldehyde, acetaldehyde and acrolein were determined at three engine operation conditions (cold idle, full power and hot idle) for five fuels. The results are shown in figures 2 to 4 with error bars. These error bars are estimated based on the detection limit of the FTIR (~2ppm). All three aldehydes emissions are at the highest levels at cold idle and the lowest levels at full power, which were directly related to the combustion temperatures.

At idle conditions, the HEFA fuels F and I showed the similar formaldehyde emissions with Jet A-1 while neat GTL fuel E and 10% FAE fuel H had lower levels of formaldehyde emissions than Jet A-1. This could be attributed to the differences in their carbon chain structure. HEFA fuels have more branched hydrocarbons than GTL fuel and similar to Jet A-1. These branched hydrocarbons have a stronger tendency for α scissions of carbon chains than straight chain hydrocarbons such as GTL fuels, which is a major formation route for formaldehyde.

At the full power condition, all the fuels had similar formaldehyde emissions with Jet A-1 except fuel H which showed higher formaldehyde emissions than the Jet A-1 value. For the FAE blend fuel H, there are two contrary factors affecting its combustion and emissions compared to Jet A-1: fuel born oxygen as a positive factor which could improve combustion and reduce emissions, and higher viscosity and lower volatility relative to Jet A-1 as negative factors which could deteriorate combustion and emissions. The higher than Jet A-1 formaldehyde emissions at full engine power for the FAE blend fuel H could be due to the fact that the negative factors (higher viscosity-approximately doubled compared to Jet A-1, and lower volatility) became dominant and affected fuel vaporisation and atomisation in the combustion chamber and thus produced higher emissions than Jet A-1. At the cold and hot idle conditions, the positive factor (fuel born oxygen) became dominant and assisted the oxidation of the fuel. As a result, formaldehyde emissions from the fuel H were lower than the Jet A-1 values at idle. The overall effect of the positive and negative factors on the combustion and emissions is related to engine load and ultimately fuel flow. At the idle conditions, the fuel flow rate to the engine was low and the impact of viscosity and volatility was low. Therefore the positive effect of fuel born oxygen became dominant. At the full power condition, as the fuel flow rate to the engine increased significantly, the impact of inferior vaporisation and atomisation properties of FAE became significant and thus the combustion efficiency deteriorated compared to Jet A-1. Pandey et al ([Pandey et al., 2012](#_ENREF_22)) analysed the correlation of biodiesel’s properties and fuel spray characteristics in diesel engines and found that viscosity of fuels could have great influences on atomization and vaporization, which could therefore affect combustion efficiency and emissions. Szybist ([Szybist et al., 2007](#_ENREF_27)) and Habib ([Habib et al., 2010](#_ENREF_8)) showed that the presence of oxygen in the biodiesel can reduce particulate matter and CO emissions, which supported the findings that the presence of oxygen in the FAE could reduce formaldehyde emissions.

By the comparison of the cold and hot idle emissions, it was observed that the engine produced higher formaldehyde emissions at cold idle than that at hot idle, indicating a lower combustion temperature at cold idle due to the cold engine components. The emission index of formaldehyde for Jet A-1 is ~4.5 g/kg-fuel and 3.6 g/kg-fuel at cold and hot idles respectively, much higher than the values measured by Knighton et al ([Knighton et al., 2007a](#_ENREF_13)) from CMF56-2-C1 engines of a DC-8 aircraft in APEX program, in which they reported approximately 0.300~2.5 g/kg-fuel at 4% rated thrust (ground idle condition). The large variations in Knighton’s data were due to the change of ambient temperatures. The measurement in this research was taken via the indoor test and therefore the effect of ambient temperatures was minimal. Spicer et al ([Spicer et al., 1999](#_ENREF_26)) measured formaldehyde from aircraft gas turbine engines and reported a value of 1.24 g/kg-fuel at 4% rated thrust, one third of the hot idle formaldehyde emissions of this research. The high aldehydes emissions from this research are due to the engine technology, as the engine used in this paper is 1950’s design whereas Knighton et al used a modern engine.

The formaldehyde emissions at cold idle were two to threefold higher than that at full power for Jet A-1 and two HEFA blends (fuels F and I). For the FAE blend (fuel H), the difference between idle and full power was small due to its relatively lower idle emissions and higher full power emissions. Wei et al ([Wei et al., 2009](#_ENREF_28)) investigated the aldehydes emission mechanisms from blend fuels using the spark ignition engine. They concluded that the emissions of formaldehyde and acetaldehyde are controlled by combustion temperature and gas flow velocity (i.e. residence time). No formaldehyde and acetaldehyde will be emitted when the temperature is too high or too low due to rapid consumption and the frozen generation respectively. The high formaldehyde emissions at idle in this study were due to relatively low combustion temperatures and thus low consumption that would be accelerated at higher temperatures. The fuel borne oxygen accelerated the consumption of formaldehyde at the idle condition with the FAE blend (fuel H), thus lowering the formaldehyde concentration.

Figure 2 Formaldehyde emissions for each fuel at three different engine operation conditions

Though acetaldehyde and acrolein emissions, as shown in figures 3 and 4, had a general trend of the highest at cold idle and lower at hot idle and the lowest at full power, similar to the trend of formaldehyde emissions, the error bars for these two aldehydes are large, indicating a greater uncertainty for the values due to the sensitivity of the FTIR. Therefore the reductions at hot idle was marginal. However, the reductions at full power are still evident for acetaldehyde and acrolein. Figure 4 shows the acrolein emission index for all fuels. The differences were not discernible between cold and hot idles. Notable reductions at full power were observed.

Figure 3 Acetaldehyde emissions for each fuel at three different engine operation conditions

Figure 4 Acrolein emissions for each fuel at three different engine operation conditions

Formaldehyde, acetaldehyde and acrolein emissions were summated as the total aldehyde emissions in terms of mg/kg-fuel. The fractions of each aldehyde as a ratio of total aldehydes emissions are presented in figure 5. The results show that formaldehyde is dominant for all conditions and fuels with around 60% of share except fuel H at full power. Acrolein as the second dominant aldehyde has a share of around 30%. Acetaldehyde emissions were very low for all the fuels and have around 10% of total aldehydes. The uncertainty for the acetaldehyde emission was high due to that its concentrations were close to the detection limit of the FITR and there were no clear differentiations between fuels. The dominance of formaldehyde could be due to two reasons: the breakup of hydrocarbon chains during the partial oxidation of fuel is mainly α scissions of carbon chains and that formaldehyde is more stable and its bond dissociation energy is much higher than other aldehydes.

Figure 5 Fractions of formaldehyde, acetaldehyde and acrolein in total measured aldehyde emissions (EI) at three engine operation conditions for different fuels

## 3.2 Comparison with Jet A-1

One of the criteria for assessing any aviation alternative fuels is their performance relative to conventional Jet A-1. It is hoped that alternative fuels provide improved performance including emissions or at least not worse than Jet A-1. Thus aldehydes emissions from four alternative/renewable fuels and their blends were normalised to Jet A-1 as shown in figures 6-8.

Figure 6 shows the normalised formaldehyde emissions. The reductions of 30~40% were observed for FAE fuel blend H and GTL fuel E at idle compared to Jet A-1. Fuel H showed a 43% increase at full power compared to Jet A-1. The rest of formaldehyde emissions showed ±5% variations relative to Jet A-1.

Figure 6 Comparison of formaldehyde emissions normalized to Jet A-1 at three engine operation conditions

The normalised acetaldehyde emissions in figure 7 show there are no obvious differences between different fuels. The large error bars at full power were due to that the concentrations of acetaldehyde in the exhaust at full power were close to the FTIR detection limit.

The normalised acrolein emissions are shown in figure 8. The results show that there are no clear differences between Jet A-1 and HEFA fuel blends (fuel F and I). Fuel H had a marginal reduction in acrolein emissions at full power compared to Jet A-1. The neat GTL fuel E showed notable reductions at cold idle and full power; particularly at cold idle with a 50±14% reduction.

Figure 7 Comparison of acetaldehyde emissions normalized to Jet A-1 at three engine operation conditions

Figure 8 Comparison of acrolein emissions normalized to Jet A-1 at three engine operation conditions

## 3.3 Fractions of formaldehyde in total hydrocarbon emissions

For aircraft engine emission regulation, total hydrocarbon emissions (THC) was measured by FID (Flame Ionisation Detector) and represented as methane equivalent in term of emission index (g/kg-fuel) for the LTO (Landing Take-off) cycle ([ICAO, 2008](#_ENREF_11)). This process counts all detected carbon atoms as CH4 and uses molecular weight of CH4 to calculate EI of THC including carbons from aldehydes. As formaldehyde is a prevalent aldehyde, the fraction of formaldehyde in THC by concentration (ppm) was determined as shown in figure 9. It shows that formaldehyde takes 6~10% at idle and ~20% at full power of THC concentrations. The percentages, however, have changed when converting concentrations to EI. The fractions of formaldehyde in THC in terms of EI in figure 10 showed almost doubled figures. This is because the molecular weight of formaldehyde (30) is almost double of methane (16).

The significance of getting fractions of formaldehyde from THC in terms of EI is that it enables estimation of formaldehyde emissions and thus ozone forming potentials (OFP) by legislated LTO THC values from ICAO database ([ICAO](#_ENREF_10)) as formaldehyde is not a directly measured pollutant.

Figure 9 Fraction of formaldehyde in total hydrocarbon emissions in term of ppm

Figure 10 Fraction of formaldehyde (g/kgfuel) in total hydrocarbon emissions (g/kgfuel, CH4 equivalent)

# CONCLUSIONS

Formaldehyde, acetaldehyde and acrolein emissions as important HAPs identified by ICAO and EPA of the USA have been investigated for four alternative aviation fuel blends and Jet A-1 using a gas turbine engine. The work has provided the knowledge and information for the development of emissions inventories in terms of aldehyde emissions, which will help the promotion of deployment of alternative fuels in aviation sector and the quantification of their environmental impacts. Overall, all four alternative fuels/blends showed equivalent or lower aldehyde emissions compared to Jet A-1. Formaldehyde appeared to be the dominant aldehyde species. The detailed conclusions are as follow:

1. Two HEFA/Jet A-1 fuel blends (fuels F and I) had similar emissions for all three aldehydes at three engine operational modes with Jet A-1.
2. The FAE blend (fuel H) showed decreased values at idle and increased values at full power for formaldehyde emissions. The contradictory effect of the FAE blend between idle (lower than Jet A-1) and full power (higher than Jet A-1) could be due to the trade-off between positive effect (improved oxidation by fuel born oxygen) and negative effect (deteriorated atomisation caused by higher viscosity and low volatility) of FAE.
3. Neat GTL fuel achieved notable reductions in formaldehyde (~30%) and acrolein (50±14%) at idle compared to Jet A-1. The lower formaldehyde emissions could be because of it lower tendency for α scissions due to its straight carbon chain. Overall formaldehyde emissions were 2~3 times higher at idle than that at full power.
4. Though the fraction of formaldehyde in THC is only 6~10% at idle and ~20% at full power, the mass emission (g/kg-fuel) of formaldehyde is doubled due to its higher molecular weight compared to methane. These fraction values can be used to estimate OFPs from legislated LTO cycle THC emission indices of aircraft engines, which are available from ICAO database with open access and thus provide a convenient way for assessing OFPs and their impact on air quality in the vicinity of airports.

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