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DC-8-based observations of aircraft CO, CH_4, N_2O, and $H_2O_{(g)}$ emission indices during SUCCESS

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Abstract. We report the first measurements of CO, CH_4, N_2O, CO_2, and $H_2O_{(g)}$ in the exhaust trails of T-39, B-757, and DC-8 aircraft at cruise conditions. Emission indices (EI) derived from these in-situ measurements are presented. Results are in agreement with ground-based tests indicating aircraft act as a net sink for CH_4 and recent airborne in-situ measurements that N_2O is not an important exhaust constituent. Condensation of $H_2O_{(g)}$ on exhaust particles resulted in EI($H_2O_{(g)}$) values less than those expected from the combustion of fuel alone. Observed apparent negative EI($H_2O_{(g)}$) values suggest that aircraft aerosol emissions, under unique atmospheric conditions, seed cloud formation and lead to dehydration of the exhaust-influenced air parcel. Such conditions may induce the formation of cirrus clouds from persistent contrails. Comparisons with the Boeing EMIT Code show measurement-derived CO emission index values consistent with model evaluations.

Introduction

Engine exhaust emissions of gases and particles from fleets of subsonic aircraft, operating primarily in the upper troposphere and lower stratosphere, are potentially in sufficient amounts to affect atmospheric ozone and climate [NASA, 1997]. Until recently, the only source-strength information available for assessing the environmental impact of these emissions has come from ground-based testing of aircraft engines in indoor facilities. It is uncertain, however, whether emission indices (EI) derived from these data are representative of aircraft operating at cruise altitude (9-13km). Thus, with fuel usage by scheduled airliner and cargo aircraft projected to triple by 2015 [Baughcum et al., 1994], increasing emissions by approximately 220% from 1990 levels [Stolarski and Wesoky, 1993], there is a growing need for in-situ measurements of aircraft effluents under actual operating conditions.

The primary gaseous emissions from aircraft engines are carbon dioxide (CO_2) and water vapor ($H_2O_{(g)}$) produced by the combustion of jet fuel. Carbon monoxide (CO) and methane (CH_4) are also produced in the combustors and vary in quantity according to the temperature, pressure, and other combustor conditions [Baughcum et al., 1996]. The emission of nitrous oxide (N_2O) is likely small from turbine combustors [Broderick et al., 1975; Zheng et al., 1994; Fahey et al., 1995b]. Since CO_2, $H_2O_{(g)}$, CH_4, and N_2O are noted greenhouse gases, and the oxidation of CO serves as a major sink for OH, quantifying the amount of these species present in aircraft exhaust is paramount to the understanding of the effect of aviation upon local, regional, and global atmospheric processes. Presented here are observations of CO_2, $H_2O_{(g)}$, CO, CH_4, and N_2O made within the exhaust plumes of three different aircraft at cruise conditions and the resulting emission indices calculated from these measurements.

Experimental

The Subsonic Aircraft: Contrail and Cloud Effects Special Study (SUCCESS) experiment was an airborne campaign based out of Salina, Kansas during the spring of 1996. NASA's DC-8 aircraft served as the primary sampling platform while their T-39 and Boeing 757 (B-757) aircraft provided the emissions source for the DC-8 target aircraft. Standard Jet A fuel was burned by all three aircraft during the experiment. On a few flights, the B-757 switched in-flight between fuel tanks containing Jet A fuel having either a high or low sulfur content (675 vs.75 ppm S).

Measurements of CO, CH_4, and N_2O were made using a three-channel, mid-IR diode laser-based differential absorption instrument while CO_2 was measured with a modified Li-Cor model 6252 non-dispersive infrared detector. Sampling of these trace species occurred through a common inlet and associated plumbing. Calibrations were performed at 10 min. intervals with measurement accuracy closely tied to the primary calibration standards obtained from the National Oceanic and Atmospheric Administration/Climate Monitoring and Diagnostics Laboratory. Data were recorded and archived at 1Hz with precisions (1σ) for CO, CH_4, N_2O and CO_2 of 1 ppbv, 1 ppbv, 0.2 ppbv and 50 ppbv respectively. A detailed discussion of the operation of these instruments during previous aircraft measurement campaigns is given by Sachse et al. [1991] and Anderson et al. [1993].

Water vapor ($H_2O_{(g)}$) was measured utilizing a fast response (~50 msec) near-IR diode laser (1.4μm) sensor recently developed [Collins et al., 1995] and flown on two missions prior to SUCCESS. This sensor is comprised of a compact laser transceiver mounted to a DC-8 window plate and a sheet of high-grade retroreflecting material that is applied to an outboard DC-8 engine housing. Using differential absorption detection techniques, $H_2O_{(g)}$ is sensed along this 28.5 m external path. An algorithm calculates $H_2O_{(g)}$

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Table 1. Flight parameters for plume measurements during SUCCESS

Emissions Source (engine)	Date (UTC)	Pressure Altitude (km)	Separation (km)	Mach #	Fuel Flow* ($lbhr^{-1}$)
T-39	960418	9.5	2.4 -12.7	0.66 - 0.76	~1000
(JT-12A-8A)	960427	9.3 - 10.7	0.3 - 10.4	0.60 - 0.78	~1000
DC-8	960429	12.5	self-exhaust	0.70 - 0.73	2500
(CFM56-2-C1)					
B-757	960503	9.0 - 11.6	4.4 - 11.9	0.59 - 0.71	2515 - 3510
(RB211-535E4)	960504	11.5 - 12.2	6.2 - 18.7	0.71 - 0.72	2375 - 2713
	960507	11.0 - 11.8	9.5 - 44.1	0.70 - 0.74	2700 - 2869
	S Jet A**	11.2	17.0 - 32.0	0.70	2803

*Fuel flow rate is per engine.
**S Jet A (Standard Jet A fuel with no added sulfur) case is for a portion of the flight on 960507 UTC.

concentration based on the differential absorption signal magnitude, ambient pressure and temperature, and spectroscopic parameters that are measured in the laboratory. The precision of the instrument is 2% of the mixing ratio. SUCCESS H_2O(g) data were recorded at 20 Hz and archived at 1 Hz.

Five flights were conducted during SUCCESS with the primary objective of exhaust sampling. Measurements of the DC-8's own exhaust were also made (self-exhaust). During the majority of the time, meteorological conditions were such that visible contrails formed. Flight parameters during exhaust sampling are given in Table 1.

Results and Discussion

The production of CO_2 from the combustion of fuel is directly related to fuel consumption. Measurements made simultaneously with CO_2 can therefore be expressed as EI which are defined as grams of species emitted per kilogram of fuel burned. Analysis of the hydrogen content of the SUCCESS Jet A fuel by the Aerospace Fuels Laboratory at Kelly AFB yielded 13.7 % hydrogen by weight and resulted in a calculated EI(CO_2) of 3159 g (kg fuel)$^{-1}$ assuming complete oxidation. For a species of interest such as CO, EI(CO) = EI(CO_2) * 28/44 * [ΔCO] / [ΔCO_2]. Multiplication by the ratio of atomic mass units is required to convert from volume to weight fractions. Values for the ratio, $\Delta CO/\Delta CO_2$, are obtained using linear regression techniques on time series of individual plume crossings. Emission indices were calculated only for time series producing a linear correlation coefficient of $r \geq 0.70$. Error bars for each EI were determined using the error of the slope and are given as upper limits. The errors are driven by the number of data points comprising the peak, peak size and shape, and variability of the background level.

Table 2 presents emission indices for all three aircraft. Due to the changing flight parameters, mean EI values are not indicated. The EI(CO) values for the T-39 are considerably larger than those for the DC-8 or B-757 reflecting the older combustor technology of that aircraft's engines. For comparison, recent in-situ emission measurements of NASA's ER-2 aircraft, which is powered by an older vintage J75 engine, revealed EI(CO) values of 18.3 to 20.2 g kg^{-1} [Fahey et al., 1995b]. Despite these comparatively large CO emissions, it should be noted that only a few T-39 aircraft are in operation today whereas DC-8 and B-757 (~ 422 out of 748 B-757's have RB211 engines) aircraft are more commonplace in the current global fleet. Median values for the B-757 are below those of the DC-8 on all three days of in-situ sampling and indicative of the newer engine technology on that aircraft. Variability in measurements behind the same aircraft can be attributed to the dependence of EI(CO) on ambient pressure, ambient temperature, ambient relative humidity, Mach number and fuel flow rate. As shown in Table 1, flight parameters were quite variable during any given flight. CO emissions are highest at low power settings where the temperature of the engine is low and incomplete combustion occurs and they decrease with increasing fuel flow [Baughcum et al., 1994]. As an example, the wide range of EI(CO) values for the B-757 on 960503 UTC result from a change in fuel flow rate (3523 to 2815 $lbhr^{-1}$), Mach number (0.68 to 0.59), and engine gas temperature (576^{0}C to 499^0). Conversely, the compactness of the data on 960507 UTC (standard Jet A only portion) results from the reasonably constant Mach number, fuel flow rate and flight level.

Changes in CH_4 from background were observed on a number of T-39 and B-757 plume crossings and found to be anticorrelated with CO_2 in 81% of those cases suggesting the engines of these aircraft actually burn a fraction of the CH_4 contained in background air. The derived values for EI(CH_4) are noted in Table 2 where emission indices for a given

Table 2. Emission indices and statistics for CO and CH_4

	range	median	n	EI ± % (1σ)
T-39	-1.08 to -0.37*	-0.52	10	20
960418	15.94 - 18.67	17.10	10	10
960427	15.60 - 26.55	20.98	72	5
DC-8				
960429	2.06 - 3.43	2.53	6	20
B-757	-1.60 to 1.19*	-0.02	6	20
960503	1.02 - 4.45	1.79	39	13
960504	0.84 - 3.86	1.55	15	20
960507	0.77 - 1.86	1.16	8	14

*EI(CH_4) values.
n = sample size.

aircraft have been combined for different days due to the small sample size. The low number of EI(CH_4) determinations can be attributed to fewer crossings having a correlation coefficient $r \geq 0.70$ for $\Delta CH_4/\Delta CO_2$. Since EI(CH_4) is small, plume dilution can lead to no detectable change in CH_4 [Zheng et al., 1994]. All detectable measurements behind the T-39 were found to be depleted in CH_4 relative to the background concentration. Peak ΔCH_4 values for these cases ranged from -5.8 to -26 ppbv. These results are consistent with the findings of Spicer et al. [1992] who reported ground-based testing of F101 and F110 engine exhaust depleted in CH_4 compared with incoming air used for combustion at power levels above idle. Similarly, Wiesen et al. [1994] observed decreasing CH4 concentrations with increasing engine thrust in the ground-based testing of the PW 305 engine. For the B-757, CH_4 was depleted in three cases (ΔCH_4 = -8.7 to -7.5 ppbv) and enhanced in three (ΔCH_4 =7 to 19.8 ppbv). CH4 production occurred during periods of changing engine thrust conditions. The potential for some CH_4 production during combustion was noted by Spicer et al. [1992]. Altitude test chamber measurements of RB211 (e.g. B-757) engine exhaust showed no changes in CH_4 concentrations [Wiesen et al., 1994].

N_2O was found not to be an important exhaust constituent which is in agreement with in-situ measurement results reported by Zheng [1994] and Fahey [1995b]. In contrast, altitude chamber tests of the RB211 engine showed N_2O concentrations tending to increase with increasing engine power however, combustor inlet temperatures were much higher than under normal cruise conditions [Wiesen et al., 1994].

Based on the hydrogen content analysis of the fuel and assuming 100% combustion efficiency, an emission index for water of 1228 g (kg fuel)$^{-1}$ is expected. However, the apparent emission indices observed for $H_2O_{(g)}$ (EI($H_2O_{(g)}$)) during SUCCESS were often substantially different from this value. This was expected, due in part to the different sampling volumes viewed by the CO_2 and $H_2O_{(g)}$ instruments and to the vertical displacement of the plumes by dynamical/thermodynamical effects, but primarily from the formation of condensation trails in the aircraft wakes. Regarding sampling error, aircraft exhaust plumes exhibit large spatial variability in species concentration so that single point measurements (e.g. CO2 data) will seldom correspond exactly to concentrations averaged over 10's of meters of path length (e.g. the $H2O_{(g)}$ data). In addition, the effects of wingtip vortices and exhaust buoyancy combine to displace aircraft plumes several tens of meters vertically from their original deposition altitude [Miake-Lye et al., 1993a]. Because EI are calculated from the species enhancements relative to background air and water vapor concentrations frequently show steep vertical gradients in the upper troposphere, this effect can introduce a significant error in EI determinations for this species. However, the large deviations of EI($H2O_{(g)}$) from the predicted value occurred when contrails were formed. In these cases, EI($H2O_{(g)}$) was less than predicted and indeed, under unique atmospheric conditions, exhibited apparent negative values, which indicates contrail growth took place at the expense of water vapor from the background atmosphere. Cases illustrating this effect are discussed below.

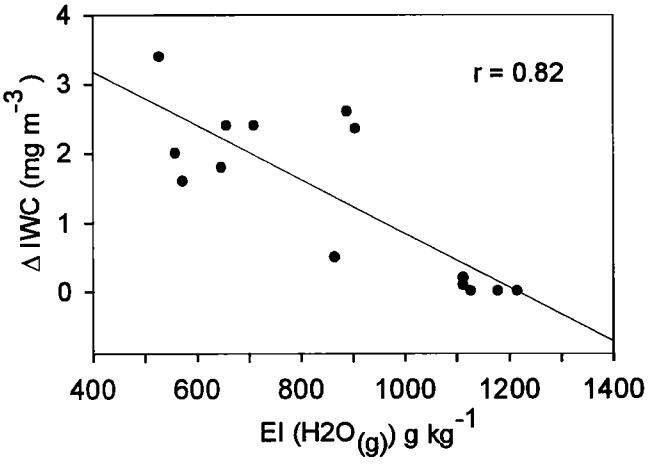

Figure 1. 960507 DC-8 chasing the B-757 which is burning standard Jet A fuel. EI($H_2O_{(g)}$) at or near 1228 g kg^{-1} are for plume sampling with no contrail formation.

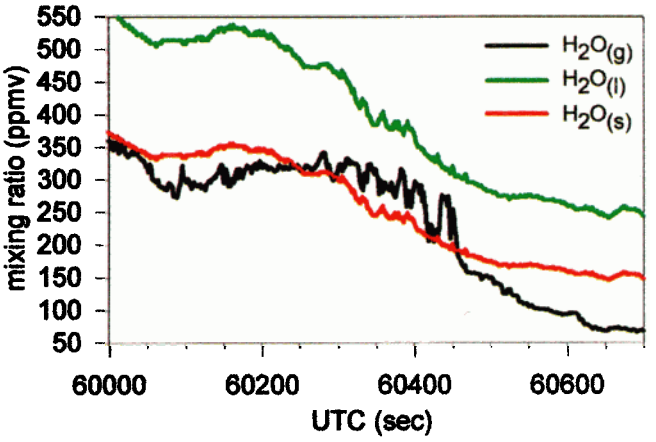

Figure 2. Mixing ratios for saturation with respect to $H_2O_{(l)}$, saturation with respect to $H_2O_{(s)}$, and ambient $H_2O_{(g)}$ levels during periods of observed anticorrelation of $H_2O_{(g)}$ relative to CO_2 between 60326 - 60460 UTC on 960427.

Figure 1 depicts EI($H_2O_{(g)}$) as a function of the change in cloud ice water content (ΔIWC) for a flight behind the B-757 where standard Jet A fuel was being burned and illustrates the inverse relationship observed between ΔIWC and EI($H_2O_{(g)}$). Times when sampling the plume, but with no contrail formation, are reflected by the values at or near 1228 g (kg fuel)$^{-1}$. Lower than unity EI($H_2O_{(g)}$) values (i.e. EI($H_2O_{(g)}$) /1228) were realized for both B-757 low and higher sulfur fuel cases. No correlation between the sulfur content of the fuel and the degree below unity for EI($H_2O_{(g)}$) was observed.

Figure 2 shows a time series of $H_2O_{(g)}$ measurements recorded during a period when this species was anticorrelated with CO_2 in crossings of the T-39 plume and thus producing apparent negative EI($H_2O_{(g)}$) values. Near-field (< 3 km separation) samples of the aircraft emissions recorded between 60326-60460 UTC, a time during which the T-39 was producing a persistent contrail, all exhibited a depletion in $H_2O_{(g)}$ relative to background concentrations. Indeed apparent values for EI($H_2O_{(g)}$) ranged from -2391 to -4911 g (kg fuel)$^{-1}$ for the three crossings recorded. During this

period, the static air temperature was $\leq$-44.5°C and the ambient $H_2O_{(g)}$ mixing ratio was less than that calculated for saturation relative to $H_2O_{(l)}$ but greater than saturation relative to $H_2O_{(s)}$. Schmidt [1941] noted that contrails are expected to form when the exhaust mixture reaches liquid saturation. In this case, the background air was already greatly saturated relative to $H_2O_{(s)}$ and thus the aircraft aerosol emissions served to condense ambient water vapor and consequently dehydrate the parcel of air influenced by the aircraft exhaust plume. We suspect that such conditions exist in regions where contrails are observed to persist and eventually evolve into sheets of cirrus clouds. In such cases, EI($H_2O_{(g)}$) values actually provide a measure of the water vapor removed from the background atmosphere due to the presence of the contrail. In the examples given, this amounts to about 2 to 5 kg ($H_2O_{(g)}$) kg^{-1} fuel burned.

Model Calculations

The in-situ measurements of the effluents from aircraft during SUCCESS have offered the opportunity to compare data obtained under true flight conditions with model results. Such intercomparisons are necessary in order to ascertain whether models based on EI from ground-based testing accurately assess the impact of aircraft emissions on the atmosphere. A comparison of the EI(CO) values for the DC-8 on 960429 UTC and the B-757 on 960507 UTC was made with the Boeing EMIT code. Required model inputs for direct comparison with flight results were ambient pressure, ambient temperature, ambient humidity, Mach number, fuel flow rate, and engine type. During both periods of interest, standard Jet A fuel was being burned and reasonably constant flight conditions were experienced. Emission information on the T-39 JT-12A-8A engine was not available therefore an in-situ vs. model comparison was not possible. From the intercomparison, the in-situ measurements for the B-757 were lower than those predicted by the model differing by 13-21% while those for the DC-8 were higher by 14-30%. This is reasonably good agreement between the two methods considering some of the uncertainties involved and it increases the confidence in using such models in the assessment of the impact of aircraft emissions on the atmosphere. For example, from these model results the impact from aircraft on the future global CO budget can be estimated. The projected annual global fuel use for the passenger jet fleet in the year 2015 is 2.45 x 10^{11} kg yr^{-1} [Baughcum et al., 1994]. Assuming all future commercial aircraft have the modern engine technology of the B-757, which the above model results estimate have an EI(CO)=1.5 g kg^{-1} at cruise conditions, then the amount of CO emitted annually by the B-757 fleet becomes 3.65 x 10^{11} g CO yr^{-1}. Using a global CO emission rate of 2100 Tg yr^{-1} [Logan, 1994], the CO emissions from the B-757 commercial fleet in 2015 would comprise 1.74 x 10^{-2} % of the global budget; a clearly insignificant amount assuming the global CO emission rate is not vastly different in 2015.

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