Measurements of HONO, NO, NO_y and SO_2 in aircraft exhaust plumes at cruise

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[1] Measurements of gaseous nitrogen and sulfur oxide emissions in young aircraft exhaust plumes give insight into chemical oxidation processes inside aircraft engines. Particularly, the OH-induced formation of nitrous acid (HONO) from nitrogen oxide (NO) and sulfuric acid (H_2SO_4) from sulfur dioxide (SO_2) inside the turbine which is highly uncertain, need detailed analysis to address the climate impact of aviation. We report on airborne in situ measurements at cruise altitudes of HONO, NO, NO_y, and SO_2 in 9 wakes of 8 different types of modern jet airliners, including for the first time also an A380. Measurements of HONO and SO_2 were made with an ITCIMS (Ion Trap Chemical Ionization Mass Spectrometer) using a new ion-reaction scheme involving SF_5^- reagent ions. The measured molar ratios HONO/NO and HONO/NO_y with averages of 0.038 ± 0.010 and 0.027 ± 0.005 were found to decrease systematically with increasing NO_x emission-index (EI NO_x). We calculate an average EI HONO of 0.31 ± 0.12 g NO_2 kg^{-1}. Using reliable measurements of HONO and NO_y, which are less adhesive than H_2SO_4 to the inlet walls, we derive the OH-induced conversion fraction of fuel sulfur to sulfuric acid ε with an average of 2.2 ± 0.5 %. ε also tends to decrease with increasing EI NO_x, consistent with earlier model simulations. The lowest HONO/NO, HONO/NO_y and ε was observed for the largest passenger aircraft A380. **Citation:** Jurkat, T., C. Voigt, F. Arnold, H. Schlager, J. Kleffmann, H. Aufmhoff, D. Schäuble, M. Schaefer, and U. Schumann (2011), Measurements of HONO, NO, NO_y and SO_2 in aircraft exhaust plumes at cruise, *Geophys. Res. Lett.*, *38*, L10807, doi:10.1029/2011GL046884.

1. Introduction

[2] Aircraft engines generate numerous pollutants, which after release to the atmosphere may influence atmospheric processes. Emissions of carbon dioxide, nitrogen oxides, water, sulfur species, soot and chemi-ions modify the composition and radiative transfer of the atmosphere [*Intergovernmental Panel on Climate Change*, 1999]. Of the primary gases NO, NO_2, and SO_2 undergo OH-induced conversion to HONO, HNO_3, H_2SO_4 and hydro carbons already predominantly inside the post-combustor flow till the engine exit. HONO serves as a temporary reservoir of OH, which can be liberated by HONO photolysis in the atmosphere [*Arnold et al.*, 1992] and then may participate in formation of secondary H_2SO_4. H_2SO_4 is a precursor for radiatively active sulfate particles and takes part in soot particle coating. The amount of H_2SO_4 emitted by aircraft is therefore a key parameter to assess the impact of aviation on the earth's radiative budget. OH is produced in the engine combustor and subsequently decreased in the turbine segment till the engine exit to a level of the order of 10^{12} molecules cm^{-3} [*Tremmel et al.*, 1998]. This occurs mainly due to reaction with NO, NO_2 and SO_2 and partly due to self-reaction.

[3] The chemical evolution of NO_x in the exhaust plume has been widely studied on the ground using dedicated engine tests on a stationary aircraft [*Wormhoudt et al.*, 2007]. They reveal a significant dependence on the engine type and rated thrust. Comparability of cruise and ground measurements is limited due to a lack of airborne measurements.

[4] The total conversion fraction of fuel sulfur to sulfuric acid (ε) has been studied in the past by a number of measurements and model simulations summarized by *Katragkou et al.* [2004, and references therein]. At present values range between 2 and 10 % while the ICAO suggests an average ε of 2.4 wt %.

[5] Here, we report on an extensive data set of SO_2, HONO, NO and NO_y airborne measurements obtained with the DLR research aircraft Falcon during the CONCERT (CONtrail and Cirrus ExpeRimenT) campaign [*Voigt et al.*, 2010] in November 2008. Particularly the measurements in the exhaust plume of the A380 allow for a detailed discussion of the chemical evolution in the aging plume. We use the abundance ratio of HONO/NO_y to estimate the OH-induced conversion fraction ε of fuel sulfur to sulfuric acid in the aircraft engine for each aircraft plume encounter.

2. The ITCIMS Instrument

[6] Airborne measurements were performed at cruise altitude using an ion trap chemical ionization mass spectrometer (ITCIMS) combined with an SF_5^- ion source. The instrument described by *Jurkat et al.* [2010] and *Schmale et al.* [2010] measured HONO, SO_2 and the stratospheric tracers HCl and HNO_3 at a time resolution of approximately 1.6 s.

[7] The ITCIMS has been used previously for aircraft measurements with different reagent ions [*Roiger et al.*, 2011, and references therein]. The SF_5^- ion source consisted of a polonium 210 alpha emitter which was flushed with the source gas SCF_8 in N_2/propane. Fast ion trap sampling

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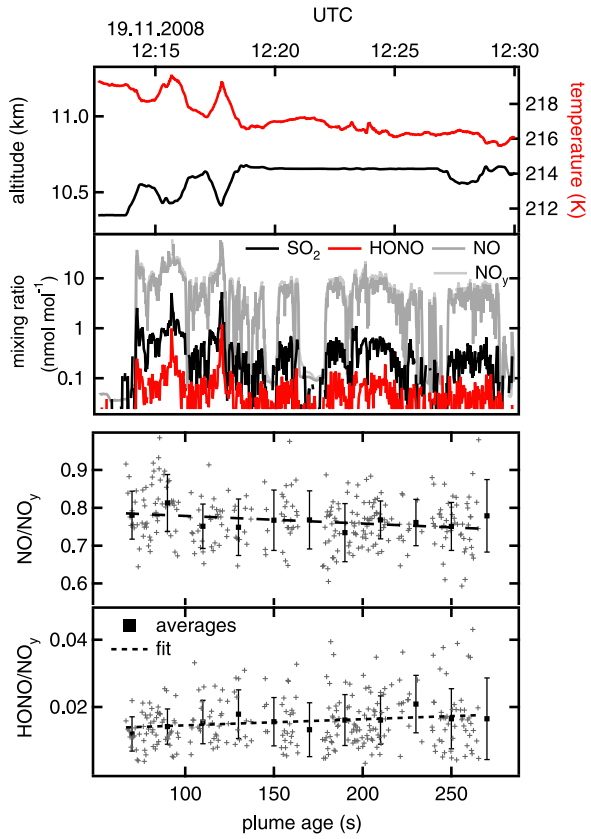

Figure 1. (top) Time sequence of the HONO, SO_2, NO and NO_y mixing ratios inside the plume of the A380 together with temperature and altitude. (bottom) HONO/NO_y and NO/NO_y versus plume age. The different plume ages evolve from a different speed of the two aircraft.

and read out proved to be particularly useful for analysis of air masses with small-scale concentration changes (background to several nmol mol^{-1} inside the center of the plume) and when many product masses were present simultaneously. After the campaign, the instrument was calibrated in the laboratory with the LOPAP (LOng Path Absorption Photometer) instrument for HONO with an accuracy of 8 % [*Kleffmann et al.*, 2006, and references therein]. The SO_2 reaction scheme and calibration are described by *Jurkat et al.* [2010] and *Schmale et al.* [2010]. The calibration factor (CF) accounts for the reaction time inside the flow reactor, the reaction rate coefficient, the dilution by the source gas flow and irreversible losses to the wall. Here, one CF for HONO and one for SO_2 were used for all flights and only applied for water vapor mixing ratios below 200 μmol mol^{-1}. This upper limit was never exceeded during the plume measurements.

[8] SO_2, NO and NO_2 may reach high concentrations in young aircraft exhaust plumes and therefore were tested on interfering reactions in the laboratory. 183 nmol mol^{-1} NO and up to 8 nmol mol^{-1} NO_2 were introduced into the upper part of the sampling line to test for interferences. Above 50 nmol mol^{-1} NO, a significant enhancement of the HONO mass peaks of a few pmol mol^{-1} was observed which can be explained by a HONO contamination of the bottled NO. NO_2 showed no interferences. Higher mixing ratios were only rarely encountered and therefore no significant interference from NO and NO_2 is expected. The estimated error of the instrument is 25 % for SO_2 and 40 % for HONO. It is dominated by the uncertainty of the relative mass discrimination of the ion trap. The detection limit (3 σ, 1.6 s) was 72 pmol mol^{-1} for HONO and 67 pmol mol^{-1} for SO_2.

[9] NO and reactive nitrogen species (NO_y) were measured simultaneously on the Falcon using the chemiluminescence technique [*Schlager et al.*, 1997]. The instrument measured NO and NO_y at a 1 Hz time resolution and an accuracy of ±8–12 % . The instrumental setup as well as a description of other instruments on the Falcon during CONCERT is given by *Voigt et al.* [2010].

3. Chemical Evolution in the Aircraft Exhaust Plumes

3.1. Aircraft Plume Sampling During CONCERT

[10] SO_2, HONO, NO, and NO_y were detected in 9 plumes from 8 different commercial aircraft, sampled during 5 Falcon flights on 17, 19 and 20 November 2008. The sampling procedure is described by *Voigt et al.* [2010]. During the chase, the pilots reported the fuel flow, engine type, Mach number, registration and weight of the aircraft. Plume ages were calculated with the Falcon wind measurements and the position of the commercial aircraft as recorded by German Air Traffic Control. The EI NO_x was calculated using the DLR fuel flow method described by *Döpelheuer and Lecht* [1999]. For the A380, the reported fuel flow of 4.75 t $(h\ engine)^{-1}$ was reduced by 30 % to match expected fuel flow records resulting in a smaller EI NO_x by about 40 %.

3.2. Plume Measurements Inside the Wake of an A380

[11] A sequence of a 13 min measurement period inside the plume of an A380 (Trent 970-87) flying at 10.65 km is exemplarily shown in Figure 1. The ITCIMS and the chemiluminescence detector show fast response times and good correlation: The highest mixing ratios of HONO of 1.2 nmol mol^{-1}, SO_2 of 5.3 nmol mol^{-1}, NO of approx. 65 nmol mol^{-1} are simultaneously encountered. The ratios of the trace gases were background corrected, the background was measured shortly before the plume encounter. The temporal evolution of the HONO/NO and NO/NO_y values for contrail ages up to 265 s of the A380 plume is also shown in Figure 1. The average ratio of 0.02 for HONO/NO stands in good agreement with ratios of *Tremmel et al.* [1998]. The average ratios of HONO/NO_y and NO/NO_y remain constant within the statistical variability of the measurements. Model simulations [*Tremmel et al.*, 1998] support the observation that titration with ozone and photolysis of HONO has a limited influence on the HONO/NO_y and NO/NO_y ratio for the measured contrail age, mainly due to low ozone mixing ratios of 40 nmol mol^{-1}.

3.3. HONO/NO and HONO/NO_y Ratios in Aircraft Exhaust Plumes

[12] Ratios of HONO/NO and HONO/NO_y for 8 different aircraft are listed in Table 1. The constant ratio of HONO/NO and HONO/NO_y with time justifies an integration over all selected data points within a given plume age interval

Table 1. Overview on the Measurements in 9 Plumes of 8 Different Aircraft[a]

Number	Engine	Aircraft-Type	FF	Weight	FL	Mach	EI NO_x	HONO/NO	$HONO/NO_y$	ε
1	CFM56-5C2	A340-311	1.25	150.3	350	0.737	11.6	0.035	0.028	0.023
2	CFM56-3C1	B737-500	(1.2)	no data	340	0.79	8.43	0.047	0.028	0.023
3	Trent556-61	A340-642	2.5	342.3	310	0.826	16.56	0.035	0.025	0.020
4a	CFM56-5B6/P	A319-111	1.15	51.1	350	0.895	11.24	0.057	0.036	0.028
4b	CFM56-5B6/P	A319-111	0.9	46.9	350	0.76	8.69	0.038	0.025	0.020
5	CF34-3B	CRJ-200	(0.52)	no data	328	no data	(9)	0.037	0.024	0.019
6	Trent970-84	A380-841	(3.6)	508.1	350	0.85	(19.69)	0.02	0.016	0.012
7	RRTayMk620-15	Fokker70	0.95	32.06	340	0.75	9.85	0.025	0.026	0.019
8	CFM56-3B1	B737-3H9	1.3	53.2	340	0.76	8.35	0.045	0.034	0.026

[a]Plume index, engine, aircraft type, fuel flow (FF, t (h engine)$^{(-1)}$, weight (t), flight level (FL), Mach number, EI NO_x (g kg^{-1}), HONO/NO, $HONO/NO_y$ and (%) ε are listed. Values in parentheses are derived with estimated fuel flow.

and dividing the integral of both species. For comparison of different aircraft plumes, the second minute past engine exit was integrated. An average ratio of 0.038 ± 0.010 for HONO/NO ratios for all plumes is obtained. $HONO/NO_y$ ratios are on average 0.029 ± 0.006 while NO/NO_y ratios range between 0.6 and 0.92 with an average of 0.72 ± 0.1. The deltas refer to a statistical variation (1 σ) of the ratios for the nine plumes. The engine specific ratios are plotted as a function of the calculated EI NO_x in Figure 2. Additionally the uncertainty regarding the EI NO_x for the A380 is included as a red horizontal bar. We observe a negative trend of the ratios of HONO/NO and $HONO/NO_y$ with increasing EI NO_x of −0.002 kg g^{-1} and −0.001 kg g^{-1}, respectively. The decrease in HONO/NO is partly caused by reaction of NO with atmospheric entrained O_3. The decrease in $HONO/NO_y$ may be attributed to OH-depletion in the high NO_x environment inside the engine or reflect elevated concentrations of organic species produced in the combustor which also react with OH. Oxidation of SO_2 and NO_2 may be affected in a similar way. Particularly the lowest value from the A380 plume may suggest that for new engines with high EI NO_x less OH is available for NO_x and SO_2 conversion.

[13] Using EI NO_x we derive the EI HONO from our measurements with an average of 0.31 ± 0.12 g NO_2 kg^{-1} which compares well to measurements behind the engine exit on the ground of *Wormhoudt et al.* [2007]. Former in-flight measurements found a ratio of 0.025 for HONO/NO in the exhaust plume of a B747 at 60 and 75 s [*Tremmel et al.*, 1998; *Arnold et al.*, 1992]. This lower limit ratio is about 40 % below the present average but agrees well with the trend at high EI NO_x. *Hanisco et al.* [1997] measured NO_y and OH in the exhaust plume of the ER-2 and the Concorde deriving a $HONO_0/NO_{y0}$ (with $HONO_0$ = HONO + HO_x and NO_{y0} at the engine exhaust plane) of 0.035 and 0.045, respectively. Our values are about a factor of 2 below their estimate, partly because we only consider the OH converted to HONO. Nevertheless, given the variability of our $HONO/NO_y$ ratios for different aircraft and the estimates discussed by *Hanisco et al.* [1997], both values agree within the measurement uncertainties.

[14] Figure 2 compares contrail to non-contrail conditions (symbolized by black squares). In general the ratios of $HONO/NO_y$ show no significant difference between contrail and non-contrail conditions. However comparison of the two plumes of the A319 (4b/4a) with and without contrail, reveals that the $HONO/NO_y$ ratio is about 30 % higher in the non-contrail plume. Heterogeneous HONO production on soot particles, OH scavenging and uptake of HONO on ice as observed for HNO_3 [*Schäuble et al.*, 2009] are thought to be of minor importance under these conditions and therefore the dominating process which may explain the difference between the two plumes remains elusive. Also, different

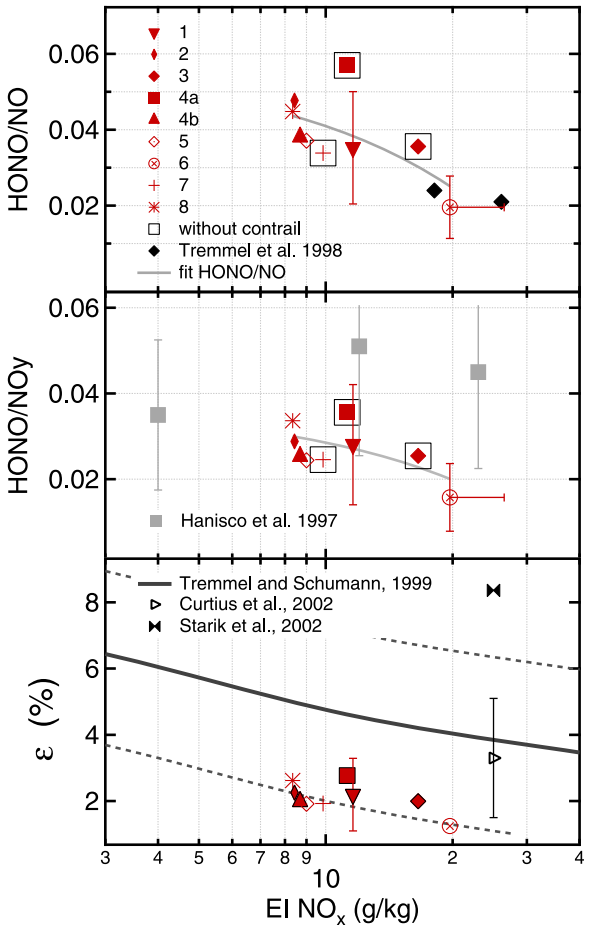

Figure 2. (top) HONO/NO and (middle) $HONO/NO_y$ are plotted versus the EI NO_x for 8 different aircraft and compared to former measurements. (bottom) The inferred sulfur conversion fraction is compared to model simulations for an upper/lower (dashed lines) and medium (thick) reaction rate coefficient. Measurements without contrails are marked with black squares. The error bars represent the total uncertainty of the measured ratios. (1) A340-311, (2) B737-500, (3) A340-642, (4 a + b) A319-111, (5) CRJ-200, (6) A380-800, (7) Fokker 70, (8) B737-3H9.

operating conditions of the aircraft (Mach number, FF, thrust etc.) may have played a role and need further research.

4. Sulfur Conversion Fraction

[15] We derive the OH-induced sulfur conversion fraction ε by comparison of the conversion fraction of NO to HONO with the rate of conversion of SO_2 to H_2SO_4. Assuming that the initial HONO/NO at the engine exit is equivalent to the $HONO/NO_y$ during our measurements and most of the OH in the post-combustor flow is consumed by the reaction with NO, we use the $HONO/NO_y$ ratio to derive an effective OH concentration. Sulfuric acid is produced in the post-combustor flow via the Stockwell-Calvert mechanism. The rate limiting step is the reaction of SO_2 with OH

$$SO_2 + OH + M \xrightarrow{k_{HSO_3}} HSO_3 + M \quad (1)$$

$$NO + OH + M \xrightarrow{k_{HONO}} HONO + M. \quad (2)$$

Assuming that most of the OH-induced conversion takes place between the combustor and the engine exit [*Starik et al.*, 2002; *Lukachko et al.*, 1998], we derive

$$[HSO_3] = \frac{k_{HSO_3}}{k_{HONO}} \frac{[HONO]}{[NO_y]} [SO_2]. \quad (3)$$

The OH-induced sulfur conversion fraction for plumes older than a few seconds is given by

$$\varepsilon = \frac{[H_2SO_4] + [SO_3]}{[SO_x]} \cong \frac{[HSO_3]}{[SO_2] + [HSO_3]} = \frac{1}{1 + \frac{[NO_y]}{\alpha[HONO]}} \quad (4)$$

with $\alpha = \frac{k_{HSO_3}}{k_{HONO}}$ being the ratio of effective rate coefficients. Thus, ε depends only on the ratio of HONO to NO_y and α. The reaction rate coefficients depend on the pressure and temperature inside the turbine segment [*Tremmel and Schumann*, 1999] where the conversion is dominated by reaction with OH. We calculated an α of 0.8 in the turbine segment of the engine behind the combustor but emphasize that α is governed by large uncertainties [*Somnitz et al.*, 2005].

[16] The values of ε for plumes of different engines and aircraft are given in Table 1 and plotted as a function of the EI NO_x in Figure 2. The CONCERT data are compared to model simulations of *Tremmel and Schumann* [1999] and *Starik et al.* [2002] and measurements of total sulfuric acid in the wake of a B757 at cruise [*Curtius et al.*, 2002]. The two dashed curves represent a lower/upper limit of the conversion fraction using minimal/maximal reaction rate coefficients ($k_{min/max} = 9.23 \cdot 10^{-14}/9.27 \cdot 10^{-13}$ $cm^3\ s^{-1}$) for the reaction of SO_2 and OH as used in their work. The ε dependence on EI NO_x was calculated for one engine (JT9D-7A) thus comparability with other engines is limited. However it is conceivable that the decrease in ε with EI NO_x is similar for most engines.

[17] An average value of 2.2 ± 0.5 % for all plumes was inferred. The decreasing trend of ε with increasing EI NO_x, mainly due to the competing reactions of NO and SO_2 with OH, is well represented.

[18] An equally large amount of SO_3 of approximately 2 % is produced from SO_2 via reaction with atomic oxygen in the combustor [*Lebedev et al.*, 2009]. Our ε therefore displays a lower limit of the total sulfur conversion fraction since production of SO_3 inside the combustor is not considered in our method. Despite large uncertainties, the inferred OH-induced conversion fraction compares reasonably well with earlier measurements and model studies of the total sulfur conversion fraction, particularly if the value for the SO_2 to SO_3 conversion in the combustor is added to our result. Additionally, our analysis provides the first extensive investigation for different engines and EI NO_x.

5. Summary and Discussion

[19] We have presented a new measurement technique for the detection of HONO and SO_2 in aircraft exhaust plumes at cruise altitude using an ion trap chemical ionization mass spectrometer with an SF_5^- ion source during the CONCERT campaign in November 2008.

[20] With this method an extensive data set of HONO/NO, $HONO/NO_y$ ratios and ε values for plumes with ages between 60 and 120 s of 8 different commercial aircraft have been collected. We infer average ratios of 0.038 and 0.027 for HONO/NO and $HONO/NO_y$, respectively and observe a negative trend of the ratios with increasing EI NO_x. Highly resolved measurements in the wake of an A380 reveal the lowest $HONO/NO_y$ ratio of 0.016. We present a new indirect method to derive the OH-induced sulfur conversion fraction ε which benefits from reliable trace gas measurements of non-sticky molecules. Furthermore the method is independent of dilution and the initial OH concentration which is highly uncertain. Our ε with an average of 2.2 ± 0.5 % lies within the range of recently published conversion fractions.

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