Atmos. Chem. Phys., 11, 13325–13337, 2011
www.atmos-chem-phys.net/11/13325/2011/
doi:10.5194/acp-11-13325-2011

Atmospheric Chemistry and Physics

Comparison of chemical characteristics of 495 biomass burning plumes intercepted by the NASA DC-8 aircraft during the ARCTAS/CARB-2008 field campaign

A. Hecobian[1,*], Z. Liu[1], C. J. Hennigan[1,], L. G. Huey[1], J. L. Jimenez[2], M. J. Cubison[2], S. Vay[3], G. S. Diskin[3], G. W. Sachse[3], A. Wisthaler[4], T. Mikoviny[4], A. J. Weinheimer[5], J. Liao[1], D. J. Knapp[5], P. O. Wennberg[6], A. Kürten[7], J. D. Crounse[6], J. St. Clair[6], Y. Wang[1], and R. J. Weber[1]**

[1]School of Earth and Atmospheric Sciences, Georgia Institute of Technology, Atlanta, GA, USA
[2]Department of Chemistry and Biochemistry and Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO, USA
[3]NASA/Langley Research Center, Hampton, VA, USA
[4]Institute for Ion Physics and Applied Physics, Innsbruck University, Innsbruck, Austria
[5]National Center for Atmospheric Research, Boulder, CO, USA
[6]Atmospheric Chemistry and Environmental Engineering, California Institute of Technology, Pasadena, CA, USA
[7]Institute for Atmospheric and Environmental Sciences, Johann Wolfgang Goethe University Frankfurt am Main, 60438 Frankfurt am Main, Germany
[*]now at: Department of Atmospheric Science, Colorado State University, Fort Collins, CO, USA
[**]now at: Center for Atmospheric Particle Studies, Carnegie Mellon University, Pittsburgh, PA, USA

Received: 6 June 2011 – Published in Atmos. Chem. Phys. Discuss.: 30 June 2011
Revised: 10 November 2011 – Accepted: 9 December 2011 – Published: 22 December 2011

Abstract. This paper compares measurements of gaseous and particulate emissions from a wide range of biomass-burning plumes intercepted by the NASA DC-8 research aircraft during the three phases of the ARCTAS-2008 experiment: ARCTAS-A, based out of Fairbanks, Alaska, USA (3 April to 19 April 2008); ARCTAS-B based out of Cold Lake, Alberta, Canada (29 June to 13 July 2008); and ARCTAS-CARB, based out of Palmdale, California, USA (18 June to 24 June 2008). Approximately 500 smoke plumes from biomass burning emissions that varied in age from minutes to days were segregated by fire source region and urban emission influences. The normalized excess mixing ratios (NEMR) of gaseous (carbon dioxide, acetonitrile, hydrogen cyanide, toluene, benzene, methane, oxides of nitrogen and ozone) and fine aerosol particulate components (nitrate, sulfate, ammonium, chloride, organic aerosols and water soluble organic carbon) of these plumes were compared. A detailed statistical analysis of the different plume categories for different gaseous and aerosol species is presented in this paper.

The comparison of NEMR values showed that CH_4 concentrations were higher in air-masses that were influenced by urban emissions. Fresh biomass burning plumes mixed with urban emissions showed a higher degree of oxidative processing in comparison with fresh biomass burning only plumes. This was evident in higher concentrations of inorganic aerosol components such as sulfate, nitrate and ammonium, but not reflected in the organic components. Lower NO_x NEMRs combined with high sulfate, nitrate and ammonium NEMRs in aerosols of plumes subject to long-range transport, when comparing all plume categories, provided evidence of advanced processing of these plumes.

Correspondence to: A. Hecobian
(arsineh@gatech.edu)

1 Introduction

Bio-fuels, prescribed burns and natural fires are sources of biomass burning smoke. Fire emissions are an important source for a wide range of atmospheric trace gases and aerosol particles that can impact biogeochemical cycles, air quality, human health, and have direct and indirect effects on the climate through influencing the global radiation budget (Crutzen et al., 1979; Crutzen and Andreae, 1990; Yamasoe et al., 2000; Guyon et al., 2003; Bein et al., 2008). Extensive

burning episodes and the persistence of these emissions in the atmosphere for weeks mean that smoke can be transported over great distances and have both regional and global impacts (LeCanut et al., 1996; Scholes and Andreae, 2000; Dickerson et al., 2002; Allen et al., 2004; Duan et al., 2004; Honrath et al., 2004; Engling et al., 2006; Fu et al., 2009). The frequency and intensity of biomass burning events and their effects are expected to be amplified in the future due to anticipated increases in global temperatures and alterations in precipitation patterns resulting from climate change (Penner et al., 1994; Narukawa et al., 1999; Reddy and Boucher, 2004; Stocks et al., 2004; Turetsky et al., 2011).

Characterization of emissions from biomass combustion has been one of the more challenging areas of atmospheric research. In recent years, many studies have been conducted to clarify the emissions and physicochemical evolution of various trace gases and aerosols from fires (e.g., Andreae et al., 2001; Decesari et al., 2006; Akagi et al., 2011). Using a variety of sampling methods, both laboratory and direct studies of fires have been used to characterize fire emissions in differing environments with various fuels and under diverse burning and meteorological conditions (Lacaux et al., 1995; Andreae and Merlet, 2001; Abel et al., 2003; Ludwig et al., 2003; Haywood et al., 2003; Iinuma et al., 2007; Cao et al., 2008; Schmidl et al., 2008; Yokelson et al., 2009; Lee et al., 2010).

The wide range of observations for different chemical species reported in ambient smoke plumes may be due to a number of reasons. Emissions of both gaseous and particulate species can vary due to fuel type (duff, pine, etc.) (Koppmann et al., 1997), fuel condition (wet/dry) (Johnson and Miyanishi, 2001), meteorological conditions in the burning region and down-wind (cloudy/clear sky/RH) (Hoffa et al., 1999), combustion phase of the fire (Gao et al., 2003) and the location and distance where the data are collected from the fires (Trentmann et al., 2003; Reid et al., 2005). Compounding the complexity of the emissions is the mixing of plumes from various regions or even the variability within the region of burning itself, resulting in the mixing of species of various chemical ages and/or species that may have been emitted under different combustion conditions. A host of physical and chemical processes in the plumes can also further complicate the analysis as the plumes age.

One way to acquire a clearer picture of the fire plumes and their evolution is the study of fire emissions from mobile airborne platforms. Analysis and presentation of data on plumes emitted from different biomass burning sources, under various ambient and transport conditions is a valuable tool in both investigating plume processing mechanisms and useful in verifying regional and global emission and dispersion model results. Here, we report on a wide range of smoke plumes and ambient fires studied during the ARCTAS (Arctic Research of the Composition of the Troposphere from Aircraft and Satellites) experiment undertaken by the National Aeronautics and Space Administration (NASA). Jacob et al. (2010) provide detailed information on the various phases of the study and the range of platforms and instrumentation deployed. Fuelberg et al. (2010) present a complete overview of detailed meteorological conditions during ARCTAS-2008, while Singh et al. (2010) illustrate the general trends of the air masses encountered during the mission.

In this paper we compare and contrast measurements of all biomass-burning plumes intercepted by the NASA DC-8 research aircraft during the three ARCTAS phases: ARCTAS-A, based out of Fairbanks, Alaska, USA (3 to 19 April 2008); ARCTAS-B, based out of Cold Lake, Alberta, Canada (29 June to 13 July 2008); and ARCTAS-CARB, based out of Palmdale, California, USA; from 18 to 24 June 2008.

The emission ratio, where the emission of a species of interest is normalized by a co-emitted, non-reactive species (typically CO or CO_2), is an important parameter used to represent fire emissions in model simulations (Helas et al., 1995; LeCanut et al., 1996; Andreae et al., 2001; Andreae and Merlet, 2001). The term emission ratio is used when sampling occurs near the fire, whereas normalized excess mixing ratios (NEMRs) are used to describe conditions after the plume has aged (Yokelson et al., 2009). NEMRs relative to CO are used in this paper where NEMR is defined as $\Delta X/\Delta CO$. X is the species of interest and ΔX (or ΔCO) is calculated by subtracting the mixing ratios of X (or CO) outside the plume from those inside the plume. Emissions from biomass burning undergo a range of chemical and physical transformations over time (Johnson and Miyanishi, 2001 and references therein). When compared to a relatively non-reacting co-emitted tracer, such as CO, primary gaseous and particulate emissions may be expected to be depleted as the plume ages due to photochemical or physical processes; whereas secondary trace gases and aerosols are expected to increase relative to CO due to production. A statistical summary of plume NEMRs is provided through contrasting various smoke emissions by broadly separating the plumes into categories according to their sources. During the ARCTAS-CARB phase of the study, where measurements focused on California emissions, extensive local wildfires provide a contrast to the boreal fires of ARCTAS-B, both in the type of material burned and the variety of other atmospheric species present in the burning region (i.e. pristine versus anthropogenic-influenced California plumes). In addition, smoke from fires that had been transported great distances and were periodically intercepted at various times during the three phases of this study, are included in the analysis. Characteristics of all the various plumes are compared, acknowledging that they often may represent very different classes of plumes due to their origins and the different processes that may have affected them during short or long transport times.

2 Experimental methods

2.1 Aircraft instrumentation

Jacob et al. (2010) provided a complete list of measurements made aboard the NASA DC-8 aircraft during ARCTAS. In this analyses the following aircraft data were used: carbon monoxide (CO), carbon dioxide (CO_2), acetonitrile (CH_3CN), hydrogen cyanide (HCN), toluene (TU), benzene (BZ), oxides of nitrogen (NO_x and NO_y), ozone (O_3), methane (CH_4), PM_1 (particulate matter with aerodynamic diameter less than 1 µm) water soluble organic carbon (WSOC), PM_1 aerosol non-refractory chemical components including sulfate (SO_4), nitrate (NO_3), ammonium (NH_4), chloride, and organic aerosol (OA). The meteorological data and aircraft position measurements (such as latitude, longitude, altitude, etc.) are also used. A list of the instruments, corresponding references to more detailed instrument descriptions and data collection rates are provided in Table 1.

To synchronize data used in the following analysis, timing of all data was checked and adjusted, if necessary, to match that of ambient water vapor (H_2O_v) concentrations. For species where measurements were performed in discrete intervals (e.g., HCN) data were interpolated over the midpoint times of the measurement for CO. Additionally, HCN and CH_3CN data were compared for all flights and adjusted for better peak alignment where values increased for both species. The data were then averaged to a 10-s timeline to obtain a uniform time-base.

2.2 Plume identification and analysis

The two main trace gases emitted from biomass burning are CO and CO_2 (Crutzen et al., 1979). To identify all burning smoke plumes in the ARCTAS data set, all flights were checked for CO and CO_2 enhancements. For this analysis, plumes were defined as an increase in concentration equivalent to twice that of the uncertainty of the measurement when the enhancement was sustained for at least 4 s. The uncertainty values for CO and CO_2 measurements were reported as 2 ppb and 0.25 ppm (Vay et al., 2011), respectively. Once the plumes were identified, CH_3CN and HCN were used as biomass burning tracers to determine if CO and CO_2 enhancements were mainly due to biomass burning or from other sources. If r^2 values for CO and CH_3CN, or CO and HCN were higher than 0.6 during the period of enhanced CO and CO_2 measurement, the plume was designated as biomass burning. To estimate background values, the measurements outside each plume were averaged and used as the background measurement for that specific plume. The duration of the background measurements were different for each plume and depended on factors such as the location of the next plume, time of flight in the same altitude as the plume, etc. From this data set, the plumes were initially separated by phase of study (ARCTAS-A, ARCTAS-B and ARCTAS-CARB). Further analyses were performed to identify the source of the smoke. This was achieved using the location of the fires (where available), the approximate transit time from the fire to the measurement, and evaluation of other emissions in the region (e.g., urban) to assess possible mixing of smoke with other emissions during transport. The details of these analyses are discussed in the following sections. The locations of the plumes are shown in Fig. 1.

2.2.1 Identification of fire source, smoke trajectory, plume age, and possible mixing with other emissions

For all the plumes identified, plume trajectory from fire to measurement point was determined through a combination of back trajectory analysis and forward plume movement estimation (using wind direction and speed). Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) analyses (http://www.arl.noaa.gov/ready/hysplit4.html) were conducted within each smoke plume, at 10 s intervals (i.e. for each data point present in the plumes), starting from the location where the aircraft first intercepted the plume. These back trajectories were extended to up to 5 days prior to the measurement. The back trajectory analyses were repeated for 3 different altitudes (the altitude where the plume was intercepted ±20 %) for each data point. Using the Fire Information for Resource Management System (FIRMS) (Justice et al., 2002; Giglio et al., 2003; Davies et al., 2009) website, a combination of the results from FLEXPART (Stohl et al., 2005) and HYSPLIT analyses were used to predict the plume's trajectory. FLEXPART (http://transport.nilu.no/flexpart) dispersion model runs for up to 6 days were used to locate the potential path of the emission from the fires to their intersection with the DC-8 flight path for each isolated plume. When both FLEXPART and HYSPLIT analyses were available, the results were compared, and if the general direction of the plume did not agree, the wind direction and speed at the location of the data collection were checked for further verification. These two methods provided similar results in all but three plumes which were excluded from further analysis. Examples of typical back trajectory analysis results for the main categories of plumes are presented in Fig. 2.

During the ARCTAS-CARB portion of the experiment, the air masses intercepted contained fire plumes emitted within or advected over somewhat rural regions with minor anthropogenic influence; on the other hand, some smoke plumes were transported over regions heavily impacted by urban emissions. These two types of cases were separated by inspecting and comparing the trajectories of the smoke plumes and trajectories of the urban emissions. If these trajectories intersected prior to the measurement, these plumes were categorized into a different class (CARB BB + Urban).

Overall, 495 aircraft intersects with biomass burning plumes from ARCTAS-A, ARCTAS-B and ARCTAS-CARB were isolated from the seventeen DC-8 flights performed

Table 1. Measurements from the NASA DC-8 Aircraft Used in the Analyses Presented in This Paper.

Measurement	Abbreviation	Data Collection Rate	References
Carbon Monoxide Water Methane	CO H_2O CH_4	1 s	Sachse et al. (1987); Diskin et al. (2002)
Carbon Dioxide	CO_2	1 s	Vay et al. (2003)
Acetonitrile Toluene Benzene	CH_3CN $C_6H_5CH_3$(TU) C_6H_6(BZ)	0.5 s	Wisthaler et al. (2002)
Hydrogen Cyanide	HCN	0.5 s	Crounse et al. (2006); Crounse et al. (2009)
Oxides of Nitrogen Ozone	NO NO_2 NO_y O_3	10 s	Weinheimer et al. (1994)
Non-refractory Submicron Aerosol Components (Sulfate, Nitrate, Ammonium, Chloride Organics)	Aerosol (SO_4, NO_3, NH_4, chloride, OA)*	1 s 10 s	DeCarlo et al. (2008)
Submicron Water Soluble Organic Carbon	WSOC	3 s	Sullivan et al. (2006)

* The symbol SO_4, rather than SO_4^{2-} is used for the aerosol sulfate measured by the AMS (and similarly for other predominantly inorganic species) because these measurements may contain contributions from organic species such as organosulfates (organonitrates, etc. for the other species) (Farmer et al., 2010).

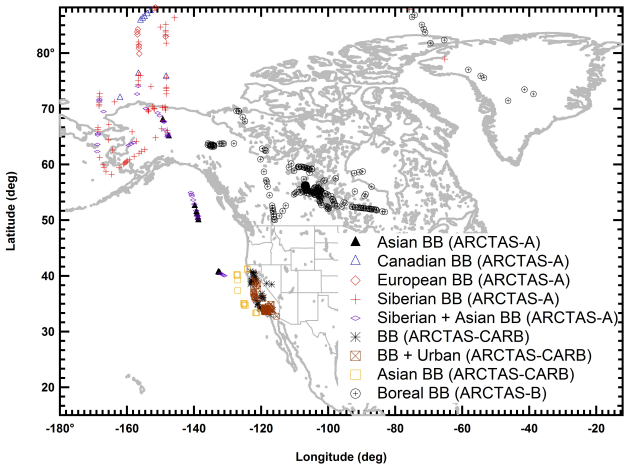

Fig. 1. Location of 495 biomass burning plumes recorded during the ARCTAS-2008 experiment aboard the DC-8 aircraft identified by fire source categories based on air mass backward trajectory analysis.

during ARCTAS-2008. The locations of the plumes are shown in Fig. 1. For some fires, there were multiple transects through what appeared to be the same smoke plume at different times and downwind distances (e.g., boreal fires). A list and description of each plume category is given in Table 2.

2.2.2 Data processing and analysis

For each smoke plume identified, NEMRs were determined relative to CO for all gaseous and aerosol components of interest (Table 1). CO has been previously used as an inert tracer for biomass burning and other emissions (e.g., Sullivan et al., 2006; Yokelson et al., 2008). The lifetime of CO in the atmosphere has been estimated to range from 1-4 months (Seinfeld and Pandis, 1998). Production of CO from VOCs in fire plumes is thought to be minor compared to the primary emitted CO concentrations. NEMRs were determined by using the mean plume concentrations and subtraction of background values, for each plume.

Table 2. Abbreviations and Description of Plumes Selected during the ARCTAS-2008 Study.

Plume Abbreviations	Plume Description
CARB BB	Biomass burning plumes that originated from California wildfires
CARB BB + Urban	Plumes of mixed California urban emissions and biomass burning plumes from California wildfires
Asian BB (CARB)	Biomass burning plumes that originated from Asian fires and were transported near the coast of California
Siberian BB (ARCTAS-A)	Plumes that were emitted from Siberian fires and encountered over the Arctic
Asian BB(ARCTAS-A)	Plumes that were originally emitted from Asian (especially Chinese) fires
European BB (ARCTAS-A)	Plumes of mixed European urban and fire emissions encountered over the Arctic
Canadian BB (ARCTAS-A)	Canadian biomass burning plumes that were encountered over the Arctic
Siberian + Asian BB (ARCTAS-A)	Mixed Asian (Chinese) and Siberian biomass burning plumes over the Arctic
Boreal Forest Fires (ARCTAS-B)	Biomass burning plumes from Canadian boreal forest wildfires

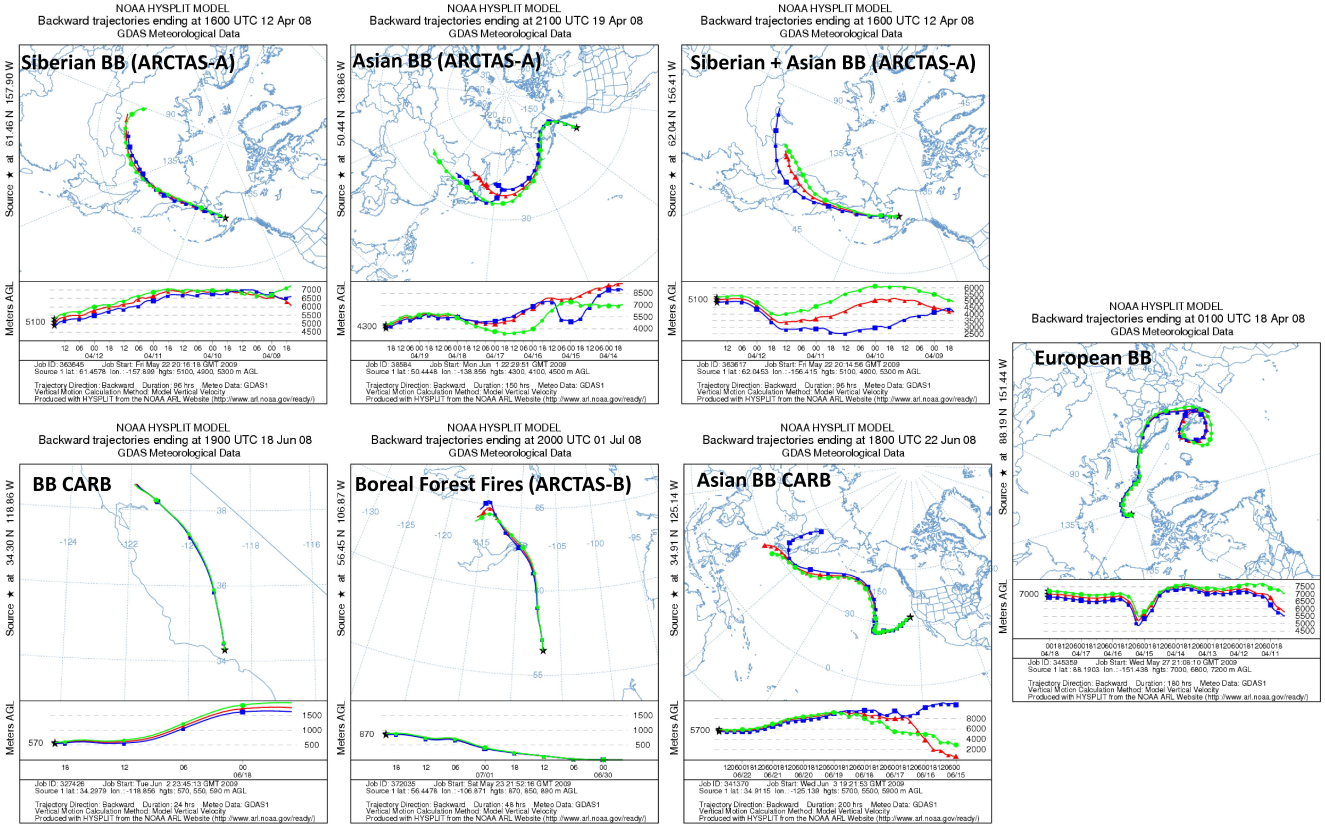

Fig. 2. Typical HYSPLIT back trajectory analysis results for some of the major plume categories.

2.3 Analyses complications and simplifications

2.3.1 Detailed data segregation versus counting statistics

The 495 plumes analyzed in this study were segregated by fire location (which is likely related to the type of material burned) and mixing with other emissions (e.g., urban). The result of this categorization is that some groups contain fewer plumes, making their statistical analysis weaker. However, the plume categorization enabled us to study emissions from different sources; this advantage is thought to override the possible statistical shortcoming.

2.3.2 Mixing of various emissions into single smoke plumes

Identifying the source of a plume, either visually when close to the fire or based on back trajectories and FIRMS data, then assuming this source is representative of a specific fire, is somewhat uncertain for a number of reasons. First, even within a single region of burning there could be differences

in material burned and burning conditions. This was visually observed in the boreal fires as areas of white and black smoke mixing within a single plume. Second, as the plume moves away from the fire, other smaller nearby fires may add fresh emissions to the plume, mixing in smoke of different ages and possibly emission characteristics. Evidence of such events was also observed in the boreal fires. Because of the averaging approach used in this analysis, these issues add uncertainty and likely some scatter to the calculated NEMRs.

2.3.3 Different losses of species relative to CO and the effect of mixing plumes of differing background concentrations

Accounting for dilution of both trace gas and aerosol particle emissions by normalizing to CO assumes that dilution is the main process leading to the loss of primary emissions as the plume moves away from the fire. This is not the case if the species in the plume experience substantial dry or wet deposition losses or uptake of gaseous species into aqueous phase. For example, soluble trace gases or hydrophilic particles are likely to be much more efficiently lost in wet scavenging events compared to CO or other insoluble/hydrophobic species. Model studies have also shown that NEMRs calculated for diluting plumes can have larger uncertainty due to background concentrations (McKeen et al., 1996). This effect is most prominent when the difference between in-plume and background concentration is low, either due to a very diffuse plume and/or a species with high background concentrations. Both of these effects are not considered here, but should mostly lead to larger relative uncertainties for the more aged and dilute plumes.

3 Results and discussion

In the following analyses, data from all fires encountered during the ARCTAS mission are compared, to contrast emission ratios in fires from a wide range of sources. This includes the boreal fires recorded in Northern Canada during ARCTAS-B discussed above, smoke plumes from springtime measurements in the Arctic (ARCTAS-A), and plumes encountered over California (ARCTAS-CARB). Although recorded in the high Arctic, practically all plumes from ARCTAS-A were associated with fires from other regions that had undergone long-range transport. For ARCTAS-CARB, most of the plumes originated from local fires, there were a only few cases of smoke transported from other regions. ARCTAS-B plumes were exclusively from boreal fires in the region of the measurements. Compared to ARCTAS-A where many plumes were subject to long range transport, plumes encountered during ARCTAS-B and ARCTAS-CARB were less aged.

During ARCTAS-A, air masses over the Arctic appeared to be broadly influenced by biomass burning emissions (Fisher et al., 2010). This was evident in the elevated HCN concentrations recorded throughout the ARCTAS-A study period. Biomass burning plumes were frequently encountered over a wide region, and generally at altitudes of $\sim 5200 \pm 800$ m a.g.l. Using HYSPLIT and FLEXPART, these biomass-burning plumes were separated into five categories based on their origin of emission. Most of the emissions were from Southern Russia (Siberian), Western China (Asian) or a mixture of both (Siberian-Asian), with a few plumes traced back to European fires. European plumes were also likely influenced by some urban emissions from cities near the burning areas. The final category of plume encountered, originated from a few fires in Canada (Canadian BB, ARCTAS-A). The number of plumes from each category is included on the bar graphs (Fig. 3a to o).

Fire emissions directly over and in the vicinity of the Central Valley were investigated during four NASA DC-8 flights on 18, 20, 22 and 24 June 2008. Also, during the return transit flight from Cold Lake to Palmdale (13 July 2008), some California fire plumes were intercepted and are included in this analysis. The average altitude of the plumes encountered for the Californian fires studied was 1500 ± 700 m a.g.l.

The California fire plumes were further separated into plumes that were influenced by urban emissions and ones that were not. Note that even for the plumes that are categorized as not influenced by urban emissions, there was always evidence for some urban influence (especially for the aged plumes); however, this influence was not as strong as the group defined here as urban-influenced (CARB BB + Urban). The correlation coefficient (r) between CO and toluene for biomass burning plumes with urban emission influence was 0.79, compared to biomass-burning plumes which were categorized as not mixed with urban emissions ($r = 0.52$). This is thought to be due to the higher variability of fire emissions compared to California urban areas. According to HYSPLIT back trajectories, these plumes traveled over areas that were less directly influenced by the large urban centers near the data collection area (e.g., Los Angeles, Sacramento, San Francisco and San Diego).

Analysis of variance (ANOVA) with Tukey's multiple range test was used to check the significance of the variation of each species when comparing different plume categories. In this analysis, the independent variables were the categories of the plumes and the dependant variables were NEMRs of gaseous and aerosol species studied. Based on $\alpha = 0.05$, all compounds show a significant difference between the means of different categories except CH_3CN, where $p = 0.2$ and WSOC where $p = 0.4$. These differences and similarities are further discussed in the following sections. A summary of the results from this analysis is presented in Table 3.

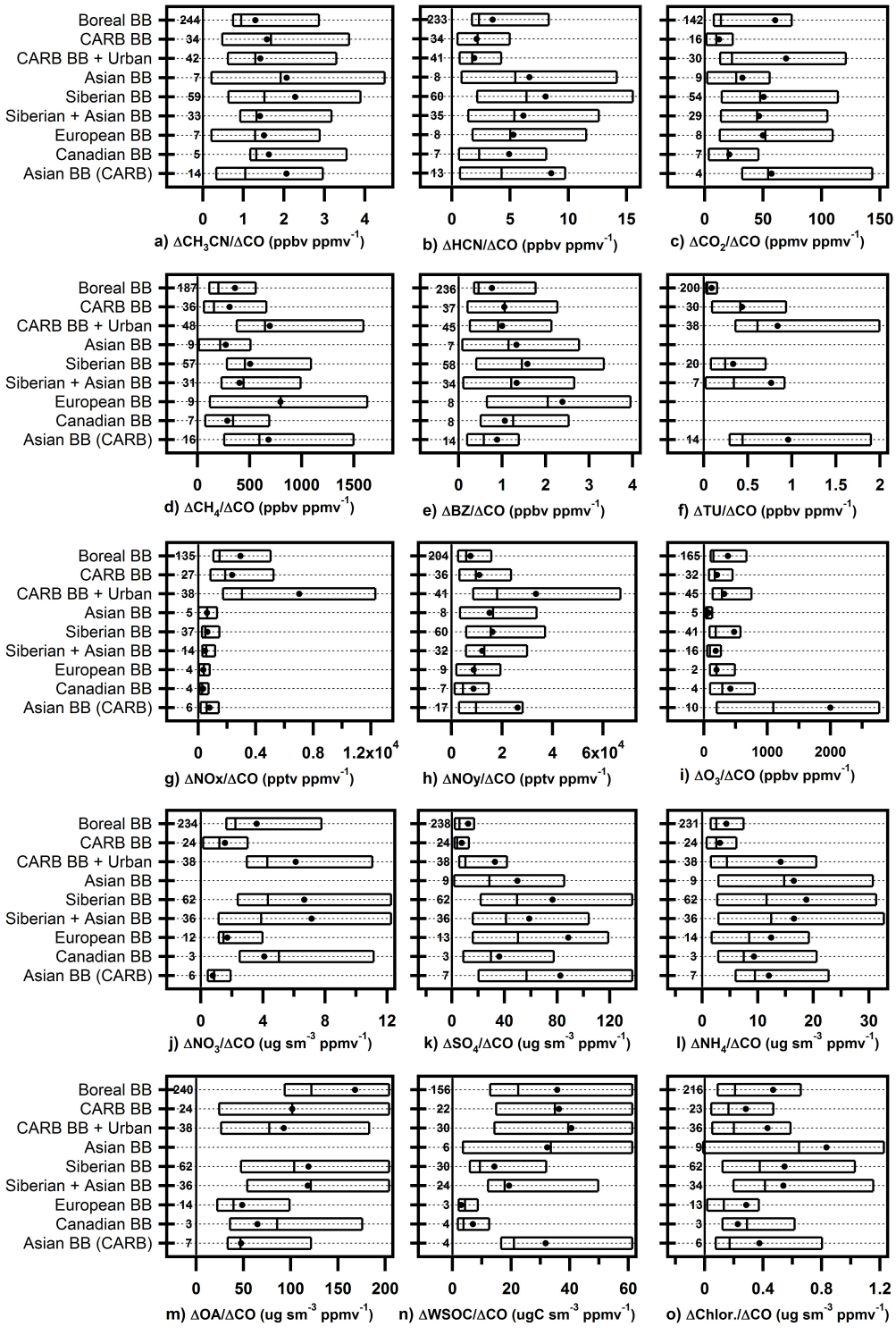

Fig. 3. Comparison of 25th and 75th percentile, mean and median NEMR values for various trace gas and aerosol components in all biomass burning plumes intercepted by the NASA DC-8 aircraft during ARCTAS-A, ARCTAS-B and ARCTAS-CARB. Numbers inside the graphs represent the number of plumes present in each category, the black dots and vertical lines in the middle are median and mean, respectively; the long black line indicates the location of zero NEMR for each compound and the sides of the rectangles are the 25th and 75th percentile values.

Table 3. Results of the statistical comparison of NEMRs of compounds in different plumes.

	Average	Std. dev	P value
$\Delta CH_3CN/\Delta CO$ ppbv ppmv^{-1}	1.7	0.4	0.4
$\Delta HCN/\Delta CO$ ppbv ppmv^{-1}	5	2	0.02
$\Delta CO_2/\Delta CO$ ppmv ppmv^{-1}	44	19	0.02
$\Delta CH_4/\Delta CO$ ppbv ppmv^{-1}	478	199	0.03
$\Delta BZ/\Delta CO$ ppbv ppmv^{-1}	1.3	0.5	0.002
$\Delta TU/\Delta CO$ ppbv ppmv^{-1}	0.6	0.3	0.03
$\Delta NO_x/\Delta CO$ pptv ppmv^{-1}	1721	2196	8×10^{-5}
$\Delta NO_y/\Delta CO$ pptv ppmv^{-1}	15 440	8820	1×10^{-6}
$\Delta O_3/\Delta CO$ ppbv ppmv^{-1}	472	585	9×10^{-6}
$\Delta NO_3/\Delta CO$ ppbv ppmv^{-1}	4	2	4×10^{-5}
$\Delta SO_4/\Delta CO$ µg sm^{-3} ppmv^{-1}	49	30	4×10^{-7}
$\Delta NH_4/\Delta CO$ µg sm^{-3} ppmv^{-1}	12	5	5×10^{-11}
$\Delta OA/\Delta CO$ µg sm^{-3} ppmv^{-1}	95	41	0.02
$\Delta WSOC/\Delta CO$ µgC sm^{-3} ppmv^{-1}	24	14	0.2
$\Delta chloride/\Delta CO$ µg sm^{-3} ppmv^{-1}	0.4	0.2	0.0005

3.1 $\Delta CH_3CN/\Delta CO$ and $\Delta HCN/\Delta CO$ (Fig. 3a and b)

Acetonitrile (CH_3CN) and hydrogen cyanide (HCN) are expected to vary little with plume age. Figure 3a shows the range of CH_3CN NEMRs and Fig. 3b presents the range of NEMRs for HCN in each plume category. The overall range for CH_3CN NEMR was 0.1–5 ppbv ppmv^{-1}. Simpson et al. (2011) reported average emission ratios of 1.8 ± 0.3 ppbv ppmv^{-1} from five Canadian boreal forest fire plumes encountered during ARCTAS-2008 for CH_3CN. Grieshop et al. (2009) recorded $\Delta CH_3CN/\Delta CO$ of 0.1–0.8 ppbv ppmv^{-1} in biomass burning simulated chamber studies. Warneke et al. (2009) reported an average of 3.1 ppbv ppmv^{-1} for agricultural fires, 2.1 ppbv ppmv^{-1} for fires from Lake Baikal and 2.4 ppbv ppmv^{-1} from Canadian boreal forest fires, while Jost et al. (2003) reported values of 3.7 and 4.1 pptv ppbv^{-1} for young Namibian biomass burning plumes. These values are consistent with the range of median values for all fires that were encountered during ARCTAS (1–2 ppbv ppmv^{-1}). Aiken et al. (2010) compare data from many studies for the observed ranges of $\Delta CH_3CN/\Delta CO$ and $\Delta HCN/\Delta CO$ in multiple laboratory and field fire studies. These values range from 0.1–4 ppbv ppmv^{-1} for CH_3CN, again consistent with the ARCTAS observations. $\Delta HCN/\Delta CO$ ratios observed here were 0.5–15 ppbv ppmv^{-1}. The observed $\Delta HCN/\Delta CO$ ratios here are also consistent with 8.2 ± 2.0 ppbv ppmv^{-1} average emission ratios reported by Simpson et al. (2011) and the range of 2–9.6 ppbv ppmv^{-1} summarized by Aiken et al. (2010).

3.2 $\Delta CO_2/\Delta CO$ and $\Delta CH_4/\Delta CO$ (Fig. 3c and d)

$\Delta CO_2/\Delta CO$ ratios were typically less than 50 to 60 ppmv ppmv^{-1}, but in some cases the distributions were skewed towards higher ratios, such as the CARB BB + Urban fire plumes. The highest CO_2 NEMRs were recorded in Asian biomass burning plumes (Asian CARB). NEMRs of methane span a large range from near zero to about 1800 ppbv ppmv^{-1} with generally higher values in the biomass burning plumes that were influenced by urban emissions (e.g., CARB BB + Urban, European BB and Asian BB (CARB)). Simpson et al. (2011) reported average emission ratios of 72 ± 44 ppbv ppmv^{-1} for methane, for the five Canadian boreal forest fire plumes encountered during ARCTAS-2008. Modified combustion efficiency (MCE), a parameter which is often used to characterize the flaming vs. smoldering nature of the fires was calculated for each category (Yokelson et al., 2008). Kondo et al. (2011) present an analysis of the biomass burning plumes encountered during ARCTAS-2008 using MCE to categorize various plumes. In the data presented here, these values ranged from 0.79 to 0.95 for the different fire categories, with higher values found in boreal fire plumes and CARB fires and lower values for fires that were transported over long distances. It is important to note that the original emission ratios and MCE values may have been changed, especially for plumes that were subject to long range transport.

3.3 $\Delta BZ/\Delta CO$ and $\Delta TU/\Delta CO$ (Fig. 3e and f)

The benzene NEMRs in all the plumes were very similar (means typically between 1 and 1.5 pptv ppbv^{-1}), except for the plumes that originated from Europe. These plumes contained approximately twice the $\Delta BZ/\Delta CO$. Overall, $\Delta BZ/\Delta CO$ ratios in this study were similar to those of Simpson et al. (2011), 1.7 ± 0.3 pptv ppbv^{-1}, Warneke et al. (2009), 1.1–1.3 pptv ppbv^{-1} and Jost et al. (2003), 0.72 and 1.2 pptv ppbv^{-1}.

Toluene NEMRs were generally less than 1 ppbv ppmv^{-1}. Warneke et al. (2009) reported 0.15 pptv ppbv^{-1} of $\Delta TU/\Delta CO$ for agricultural fires and 0.2 pptv ppbv^{-1} of $\Delta TU/\Delta CO$ for fires near Lake Baikal and Canada. Jost et al. (2003) report higher values observed over young Namibian biomass burning plumes (0.73 and 0.82 pptv ppbv^{-1}). Simpson et al. (2011) analyzed five boreal forest fire plumes and reported emission ratios of 0.67 ± 0.16 pptv ppbv^{-1}. The lower ratios observed by Warneke et al. (2009) may be due to the higher reactivity of toluene and the long transport times from Asia. The highest values of $\Delta TU/\Delta CO$ were in the ARCTAS-CARB biomass burning plumes that were heavily mixed with urban emissions. Typical urban $\Delta TU/\Delta CO$ ratios vary between 0.81 and 3.07 ppbv ppmv^{-1} (de Gouw and Warneke, 2007), higher than what was recorded in the isolated fire plumes. Muhle et al. (2007) also report elevated concentrations ($\sim$2 ppb) of toluene in ambient air

masses that were influence by forest fires in California. The Asian biomass burning plumes that were intercepted near the coast of California also showed slightly higher ratios of $\Delta TU/\Delta CO$, which may reflect contributions from urban or ship emissions in this area. A number of $\Delta TU/\Delta CO$ plume categories are not plotted due to lack of data.

3.4 $\Delta NO_x/\Delta CO$, $\Delta NO_y/\Delta CO$ and $\Delta O_3/\Delta CO$ (Fig. 3g, h and i)

NO_x NEMRs were clearly highest in the California fires that were mixed with urban emissions. NO_x is expected to be depleted relative to CO as the smoke plumes age as NO_x is converted to other compounds (e.g., PAN) over time. This is reflected in the differences between NO_x NEMRs for plumes subject to long range transport vs. fresh ones. As the Siberian, European and Asian plumes were subject to long-range transport, the low concentrations of NO_x in these plumes are to be expected. In contrast, NO_y NEMRs were fairly similar in all plumes. The CARB BB + Urban plumes had higher NO_y NEMR values, when compared to other airmasses.

For $\Delta O_3/\Delta CO$ ratios, a number of trends were observed. In the California biomass burning plumes that were mixed with urban emissions, ozone NEMRs were often (but not always) higher than the biomass plumes not mixed with urban emissions. Some studies have reported greatly enhanced O_3 when fire and urban emissions interact (Lee et al., 2008). Asian BB (ARCTAS-CARB) plumes encountered near the coast of California often had higher O_3 ratios, possibly due to a large Asian anthropogenic influence, and/or due to mixing with nearby ship plumes, which is consistent with the toluene data discussed above. Chen et al. (2005) also reported the observation of O_3 enhancement in this area, due to ship plumes in an earlier aircraft study.

3.5 Aerosol components: $\Delta NO_3/\Delta CO$, $\Delta SO_4/\Delta CO$, $\Delta NH_4/\Delta CO$ and Δchloride/ΔCO (Fig. 3j, k, l and o)

The nitrate NEMR values were generally similar in ARCTAS-CARB biomass burning plumes that were mixed with urban emissions and Siberian and Siberian-Asian plumes. Lowest concentrations were recorded in Asian BB along the coast of California, where the highest O_3 NEMRs were observed. Nitrate NEMR trends were more similar to those of NO_y than NO_x for plumes that were subject to long-range transport such as the Asian and Siberian-Asian plumes.

For sulfate, lowest ratios relative to CO for all plume sources were observed in the boreal fires and California fires that were not influenced by urban sources (CARB BB + Urban). Some studies have provided evidence for the emission of SO_2 and primary sulfate from biomass burning sources (e.g., Smith et al., 2001). The most obvious feature of the sulfate data is the much higher NEMRs in plumes subject to long-range transport. Contributions from anthropogenic SO_2 emissions in the regions of the fires (e.g., use of sulfur rich fertilizers, agricultural burning and the conversion of the released SO_2 to sulfate during long range transport) may be one reason, SO_2 emissions from the Asian fires are also thought to play a role (Kondo et al., 2011). Fine particle sulfate production in Asian anthropogenic plumes advecting to North America is well documented (Peltier et al., 2008; van Donkelaar et al., 2008; Dunlea et al., 2009) and thought to be due to the conversion of a large reservoir of SO_2 to non-volatile sulfate aerosol, which is not depleted by any precipitation scavenging on route. For this study, high sulfate NEMRs were observed in the Asian plumes intercepted near the coast of California, again possibly in part due to the influence of ship emissions along the California coast or due to the use of sulfur rich compounds in crop fertilizers and the subsequent burning of the agricultural areas.

NH_4 NEMRs were similar in boreal and California fire plumes and higher in the California fire plumes that were influenced by urban emissions and possible ammonia emissions from the nearby feedlots. For the plumes subject to long-range transport, $\Delta NH_4/\Delta CO$ variability between sources followed that of $\Delta SO_4/\Delta CO$, as expected.

The chloride NEMRs (Fig. 3o) were highest in Asian biomass burning plumes and generally higher in plumes that were subject to long-range transport. The ranges of these values were similar when comparing boreal and California biomass burning plumes.

3.6 Aerosol Components $\Delta OA/\Delta CO$, $\Delta WSOC/\Delta CO$ (Fig. 4m and n)

Organic compounds comprise the largest chemical component of fine particle in smoke from biomass burning, and secondary formation may enhance aerosol mass with plume age. The emission data have wide and overlapping variability in each transport age group, likely due to the issues discussed in previous sections.

Overall, the OA and WSOC NEMRs in plumes transported over larger distances were lower compared to smoke plumes encountered closer to the sources of fire (ARCTAS-B and CARB). In this regard, the behavior of these organic components is more similar to that of nitrate and not sulfate. It is also similar to the preferential loss of fine particle OA or WSOC relative to sulfate that has been observed in Asian plumes advected to North America (Peltier et al., 2008; Dunlea et al., 2009). In those cases it was proposed that SOA and sulfate formed close to the emission sources were scavenged by precipitation during ascent to the free troposphere, and that sulfate was regenerated on route as SO_2 survived wet scavenging, but SOA was not, perhaps due to the short lifetime of SOA precursors. Similar processes may apply to these smoke plumes.

4 Conclusions

A statistical summary of NEMRs of a wide range of gaseous and particulate species relative to CO is presented for fire plumes recorded from the NASA DC-8 aircraft as part of the ARCTAS-2008 mission. Comparisons between the wide range of fires are made with identical instrumentation and thus the observed variability is not dependent on the variations in measurement methods. This summary provides a valuable reference for comparison with previous and future studies of biomass burning emissions.

Gaseous and aerosols species emitted as primary products or produced as secondary compounds were compared. CH_3CN and HCN are considered primary biomass burning emissions and often used as tracers for such sources. HCN NEMRs showed higher variability when comparing the different plume sources. The lowest HCN concentrations were observed in boreal, CARB and CARB BB + Urban biomass burning plumes. The use of HCN as a tool to normalize and estimate other biomass burning emissions has been studied in recent years. The higher variability of HCN presented in this data may complicate such a process unless more data on the variability (dependence of burning material) and its possible change with long range transport can be collected. CO_2 and CH_4 NEMRs do not provide a clear trend; however, CH_4 concentrations are higher in biomass burning plumes that were mixed with urban emissions such as CARB BB + Urban and European BB. The benzene concentrations of European BB plumes are also higher than all the other categories, reinforcing the possible mixing of these biomass burning plumes with urban emissions. When comparing NO_x NEMRs, plumes subject to long range transport are depleted in NO_x, as is expected in well processed plumes. On the other hand, NO_y concentrations are similar in different plume categories, with some high values observed in CARB BB + Urban emissions. The highest ozone concentrations are observed in Asian BB plumes that were encountered off the coast of California. As noted before, this may be due to the transport of these plumes from regions with ship emissions.

Higher values of secondary inorganic aerosol species are observed in CARB BB + Urban air-masses, when comparing the fresh plumes (i.e. boreal BB, CARB BB and CARB BB + Urban), which may be an indication of the faster oxidation rates in these plumes. This in turn, may be a result of the high oxidative potential of the plumes in this category as evident in higher NO_x, NO_y and O_3 NEMRs. The boreal emissions have the higher organic aerosol concentrations. A similar trend is not observed in WSOC data, which may be an indication of the dominant nature of primary organic emissions from these plumes.

Acknowledgements. A. H. would like to thank the NASA DC-8 crew for their assistance in making WSOC measurements on the airplane. A. W. and T. M. acknowledge financial and logistical support from the Österreichische Forschungsförderungsgesellschaft, the Tiroler Zukunftsstifung, Armin Hansel and Tilmann D. Märk. MJC and JLJ were supported by NASA NNX08AD39G. This work was supported by the NASA Tropospheric Chemistry Program under grant number NNX08AH80G.

Edited by: K. Law

References

Abel, S. J., Haywood, J. M., Highwood, E. J., Li, J., and Buseck, P. R.: Evolution of biomass burning aerosol properties from an agricultural fire in southern Africa, Geophys. Res. Lett., 30, 1783, doi:10.1029/2003gl017342, 2003.

Aiken, A. C., de Foy, B., Wiedinmyer, C., DeCarlo, P. F., Ulbrich, I. M., Wehrli, M. N., Szidat, S., Prevot, A. S. H., Noda, J., Wacker, L., Volkamer, R., Fortner, E., Wang, J., Laskin, A., Shutthanandan, V., Zheng, J., Zhang, R., Paredes-Miranda, G., Arnott, W. P., Molina, L. T., Sosa, G., Querol, X., and Jimenez, J. L.: Mexico city aerosol analysis during MILAGRO using high resolution aerosol mass spectrometry at the urban supersite (T0) – Part 2: Analysis of the biomass burning contribution and the non-fossil carbon fraction, Atmos. Chem. Phys., 10, 5315–5341, doi:10.5194/acp-10-5315-2010, 2010.

Akagi, S. K., Yokelson, R. J., Wiedinmyer, C., Alvarado, M. J., Reid, J. S., Karl, T., Crounse, J. D., and Wennberg, P. O.: Emission factors for open and domestic biomass burning for use in atmospheric models, Atmos. Chem. Phys., 11, 4039–4072, doi:10.5194/acp-11-4039-2011, 2011.

Allen, A. G., Cardoso, A. A., and da Rocha, G. O.: Influence of sugar cane burning on aerosol soluble ion composition in Southeastern Brazil, Atmos. Environ., 38, 5025–5038, doi:10.1016/j.atmosenv.2004.06.019, 2004.

Andreae, M. O. and Merlet, P.: Emission of trace gases and aerosols from biomass burning, Global Biogeochem. Cy., 15, 955–966, 2001.

Andreae, M. O., Artaxo, P., Fischer, H., Freitas, S. R., Gregoire, J. M., Hansel, A., Hoor, P., Kormann, R., Krejci, R., Lange, L., Lelieveld, J., Lindinger, W., Longo, K., Peters, W., de Reus, M., Scheeren, B., Dias, M. A. F. S., Strom, J., van Velthoven, P. F. J., and Williams, J.: Transport of biomass burning smoke to the upper troposphere by deep convection in the equatorial region, Geophys. Res. Lett., 28, 951–954, 2001.

Bein, K. J., Zhao, Y. J., Johnston, M. V., and Wexler, A. S.: Interactions between boreal wildfire and urban emissions, J. Geophys. Res.-Atmos., 113, D07304, doi:10.1029/2007jd008910, 2008.

Cao, G. L., Zhang, X. Y., Gong, S. L., and Zheng, F. C.: Investigation on emission factors of particulate matter and gaseous pollutants from crop residue burning, J. Environ. Sci.-China, 20, 50–55, 2008.

Chen, G., Huey, L. G., Trainer, M., Nicks, D., Corbett, J. J., Ryerson, T. B., Parrish, D. D., Neuman, J. A., Nowak, J. B., Tanner, D. J., Holloway, J., Brock, C. A., Crawford, J. H., Olson, J. R., Sullivan, A., Weber, R., Schauffler, S., Donnelly, S., Atlas, E., Roberts, J., Flocke, F., Hubler, G., and Fehsenfeld, F. C.: An investigation of the chemistry of ship emis-

sion plumes during ITCT 2002, J. Geophys. Res.-Atmos., 110, D10S90, doi:10.1029/2004JD005236, 2005.

Crounse, J., McKinney, K. A., Kwan, A. J., and Wennberg, P. O.: Measurement of gas-phase hydroperoxides by chemical ionization mass spectrometry, Anal. Chem., 78, 6726–6732, doi:10.1021/ac0604235, 2006.

Crounse, J. D., DeCarlo, P. F., Blake, D. R., Emmons, L. K., Campos, T. L., Apel, E. C., Clarke, A. D., Weinheimer, A. J., McCabe, D. C., Yokelson, R. J., Jimenez, J. L., and Wennberg, P. O.: Biomass burning and urban air pollution over the Central Mexican Plateau, Atmos. Chem. Phys., 9, 4929–4944, doi:10.5194/acp-9-4929-2009, 2009.

Crutzen, P. J. and Andreae, M. O.: Biomass burning in the tropics - impact on atmospheric chemistry and biogeochemical cycles, Science, 250, 1669–1678, 1990.

Crutzen, P. J., Heidt, L. E., Krasnec, J. P., Pollock, W. H., and Seiler, W.: Biomass burning as a source of atmospheric gases CO, H_2, N_2O, NO, CH_3Cl and COS, Nature, 282, 253–256, 1979.

Davies, D. K., Ilavajhala, S., Wong, M. M., and Justice, C. O.: Fire information for resource management system: Archiving and distributing modis active fire data, IEEE T. Geosci. and Remote, 47, 72–79, doi:10.1109/Tgrs.2008.2002076, 2009.

de Gouw, J. and Warneke, C.: Measurements of volatile organic compounds in the earth's atmosphere using proton-transfer-reaction mass spectrometry, Mass Spec. Rev., 26, 223–257, doi:10.1002/mas.20119, 2007.

DeCarlo, P. F., Dunlea, E. J., Kimmel, J. R., Aiken, A. C., Sueper, D., Crounse, J., Wennberg, P. O., Emmons, L., Shinozuka, Y., Clarke, A., Zhou, J., Tomlinson, J., Collins, D. R., Knapp, D., Weinheimer, A. J., Montzka, D. D., Campos, T., and Jimenez, J. L.: Fast airborne aerosol size and chemistry measurements above Mexico City and Central Mexico during the MILAGRO campaign, Atmos. Chem. Phys., 8, 4027–4048, doi:10.5194/acp-8-4027-2008, 2008.

Decesari, S., Fuzzi, S., Facchini, M. C., Mircea, M., Emblico, L., Cavalli, F., Maenhaut, W., Chi, X., Schkolnik, G., Falkovich, A., Rudich, Y., Claeys, M., Pashynska, V., Vas, G., Kourtchev, I., Vermeylen, R., Hoffer, A., Andreae, M. O., Tagliavini, E., Moretti, F., and Artaxo, P.: Characterization of the organic composition of aerosols from Rondônia, Brazil, during the LBA-SMOCC 2002 experiment and its representation through model compounds, Atmos. Chem. Phys., 6, 375–402, doi:10.5194/acp-6-375-2006, 2006.

Dickerson, R. R., Andreae, M. O., Campos, T., Mayol-Bracero, O. L., Neususs, C., and Streets, D. G.: Analysis of black carbon and carbon monoxide observed over the Indian Ocean: Implications for emissions and photochemistry, J. Geophys. Res.-Atmos., 107, 8017, doi:10.1029/2001jd000501, 2002.

Diskin, G. S., Podolske, J. R., Sachse, G. W., and Slate, T. A.: Open-path airborne tunable diode laser hygrometer, in diode lasers and applications in atmospheric sensing, SPIE Proceedings 4817, 196–204, 2002.

Duan, F. K., Liu, X. D., Yu, T., and Cachier, H.: Identification and estimate of biomass burning contribution to the urban aerosol organic carbon concentrations in Beijing, Atmos. Environ., 38, 1275–1282, doi:10.1016/j.atmosenv.2003.11.037, 2004.

Dunlea, E. J., DeCarlo, P. F., Aiken, A. C., Kimmel, J. R., Peltier, R. E., Weber, R. J., Tomlinson, J., Collins, D. R., Shinozuka, Y., McNaughton, C. S., Howell, S. G., Clarke, A. D., Emmons, L. K., Apel, E. C., Pfister, G. G., van Donkelaar, A., Martin, R. V., Millet, D. B., Heald, C. L., and Jimenez, J. L.: Evolution of Asian aerosols during transpacific transport in INTEX-B, Atmos. Chem. Phys., 9, 7257–7287, doi:10.5194/acp-9-7257-2009, 2009.

Engling, G., Herckes, P., Kreidenweis, S. M., Malm, W. C., and Collett, J. L.: Composition of the fine organic aerosol in Yosemite National Park during the 2002 Yosemite aerosol characterization study, Atmos. Environ., 40, 2959–2972, doi:10.1016/j.atmosenv.2005.12.041, 2006.

Farmer, D. K., Matsunaga, A., Docherty, K. S., Surratt, J. D., Seinfeld, J. H., Ziemann, P. J., and Jimenez, J. L.: Response of an Aerosol Mass Spectrometer to Organonitrates and Organosulfates and implications for Atmospheric Chemistry, P. Natl. Acad. Sci. USA, 107, 6670–6675, doi:10.1073/pnas.0912340107, 2010.

Fisher, J. A., Jacob, D. J., Purdy, M. T., Kopacz, M., Le Sager, P., Carouge, C., Holmes, C. D., Yantosca, R. M., Batchelor, R. L., Strong, K., Diskin, G. S., Fuelberg, H. E., Holloway, J. S., Hyer, E. J., McMillan, W. W., Warner, J., Streets, D. G., Zhang, Q., Wang, Y., and Wu, S.: Source attribution and interannual variability of Arctic pollution in spring constrained by aircraft (ARCTAS, ARCPAC) and satellite (AIRS) observations of carbon monoxide, Atmos. Chem. Phys., 10, 977–996, doi:10.5194/acp-10-977-2010, 2010.

Fu, P. Q., Kawamura, K., and Barrie, L. A.: Photochemical and other sources of organic compounds in the Canadian high Arctic aerosol pollution during winter-spring, Environ. Sci. Tech., 43, 286–292, doi:10.1021/Es803046q, 2009.

Fuelberg, H. E., Harrigan, D. L., and Sessions, W.: A meteorological overview of the ARCTAS 2008 mission, Atmos. Chem. Phys., 10, 817–842, doi:10.5194/acp-10-817-2010, 2010.

Gao, S., Hegg, D. A., Hobbs, P. V., Kirchstetter, T. W., Magi, B. I., and Sadilek, M.: Water-soluble organic components in aerosols associated with savanna fires in southern Africa: Identification, evolution, and distribution, J. Geophys. Res.-Atmos., 108, 8491, doi:10.1029/2002jd002324, 2003.

Giglio, L., Descloitres, J., Justice, C. O., and Kaufman, Y. J.: An enhanced contextual fire detection algorithm for modis, Remote Sens. Environ., 87, 273–282, doi:10.1016/S0034-4257(03)00184-6, 2003.

Grieshop, A. P., Donahue, N. M., and Robinson, A. L.: Laboratory investigation of photochemical oxidation of organic aerosol from wood fires 2: analysis of aerosol mass spectrometer data, Atmos. Chem. Phys., 9, 2227–2240, doi:10.5194/acp-9-2227-2009, 2009.

Guyon, P., Graham, B., Beck, J., Boucher, O., Gerasopoulos, E., Mayol-Bracero, O. L., Roberts, G. C., Artaxo, P., and Andreae, M. O.: Physical properties and concentration of aerosol particles over the Amazon tropical forest during background and biomass burning conditions, Atmos. Chem. Phys., 3, 951–967, doi:10.5194/acp-3-951-2003, 2003.

Haywood, J. M., Osborne, S. R., Francis, P. N., Keil, A., Formenti, P., Andreae, M. O., and Kaye, P. H.: The mean physical and optical properties of regional haze dominated by biomass burning aerosol measured from the C-130 aircraft during SAFARI 2000, J. Geophys. Res.-Atmos., 108, 8473, doi:10.1029/2002jd002226, 2003.

Helas, G., Lobert, J., Scharffe, D., Schafer, L., Goldammer, J.,

Baudet, J., Ajavon, A. L., Ahoua, B., Lacaux, J. P., Delmas, R., and Andreae, M. O.: Airborne measurements of savanna fire emissions and the regional distribution of pyrogenic pollutants over Western Africa, J. Atmos. Chem., 22, 217–239, 1995.

Hoffa, E. A., Ward, D. E., Hao, W. M., Susott, R. A., and Wakimoto, R. H.: Seasonality of carbon emissions from biomass burning in a Zambian savanna, J. Geophys. Res.-Atmos., 104, 13841–13853, 1999.

Honrath, R. E., Owen, R. C., Val Martin, M., Reid, J. S., Lapina, K., Fialho, P., Dziobak, M. P., Kleissl, J., and Westphal, D. L.: Regional and hemispheric impacts of anthropogenic and biomass burning emissions on summertime CO and O_3 in the north Atlantic lower free troposphere, J. Geophys. Res.-Atmos., 109, D24310, doi:10.1029/2004jd005147, 2004.

Iinuma, Y., Bruggemann, E., Gnauk, T., Muller, K., Andreae, M. O., Helas, G., Parmar, R., and Herrmann, H.: Source characterization of biomass burning particles: The combustion of selected European conifers, African hardwood, savanna grass, and German and Indonesian peat, J. Geophys. Res.-Atmos., 112, D08209, doi:10.1029/2006jd007120, 2007.

Jacob, D. J., Crawford, J. H., Maring, H., Clarke, A. D., Dibb, J. E., Emmons, L. K., Ferrare, R. A., Hostetler, C. A., Russell, P. B., Singh, H. B., Thompson, A. M., Shaw, G. E., McCauley, E., Pederson, J. R., and Fisher, J. A.: The Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) mission: design, execution, and first results, Atmos. Chem. Phys., 10, 5191–5212, doi:10.5194/acp-10-5191-2010, 2010.

Johnson, E. A. and Miyanishi, K., Forest fires: Behavior and ecological effects, San Diego, CA, Academic Press, 2001.

Jost, C., Trentman, J., Sprung, D., Andreae, M., McQuaid, J., and Barjat, H.: Trace gas chemistry in a young biomass burning plume over Namibia: Observations and model simulations, J. Geophys. Res.-Atmos., 108, 8482, doi:10.1029/2002JD002431, 2003.

Justice, C. O., Giglio, L., Korontzi, S., Owens, J., Morisette, J. T., Roy, D., Descloitres, J., Alleaume, S., Petitcolin, F., and Kaufman, Y.: The MODIS fire products, Remote Sens. Environ., 83, 244–262, 2002.

Kondo, Y., Matsui, H., Moteki, N., Sahu, L., Takegawa, N., Zhao, Y., Cubison, M. J., Jimenez, J. L., Vay, S., Diskin, G. S., Anderson, B., Wisthaler, A., Mikoviny, T., Fuelberg, H. E., Blake, D. R., Huey, G., Weinheimer, A. J., Knapp, D. J., and Brune, W. H.: Emissions of black carbon, organic, and inorganic aerosols from biomass burning in North America and Asia in 2008. J. Geophys. Res., 116, D08204, doi:10.1029/2010JD015152, 2011.

Koppmann, R., Khedim, A., Rudolph, J., Poppe, D., Andreae, M. O., Helas, G., Welling, M., and Zenker, T.: Emissions of organic trace gases from savanna fires in Southern Africa during the 1992 Southern African fire atmosphere research initiative and their impact on the formation of tropospheric ozone, J. Geophys. Res.-Atmos., 102, 18879–18888, 1997.

Lacaux, J. P., Brustet, J. M., Delmas, R., Menaut, J. C., Abbadie, L., Bonsang, B., Cachier, H., Baudet, J., Andreae, M. O., and Helas, G.: Biomass burning in the tropical savannas of Ivory-Coast – an overview of the field experiment fire of savannas (FOS/DECAFE-91), J. Atmos. Chem., 22, 195–216, 1995.

LeCanut, P., Andreae, M. O., Harris, G. W., Wienhold, F. G., and Zenker, T.: Airborne studies of emissions from savanna fires in Southern Africa .1. Aerosol emissions measured with a laser optical particle counter, J. Geophys. Res.-Atmos., 101, 23615–23630, 1996.

Lee, S., Kim, H. K., Yan, B., Cobb, C. E., Hennigan, C., Nichols, S., Chamber, M., Edgerton, E. S., Jansen, J. J., Hu, Y. T., Zheng, M., Weber, R. J., and Russell, A. G.: Diagnosis of aged prescribed burning plumes impacting an urban area, Environ. Sci. Tech., 42, 1438–1444, doi:10.1021/es7023059, 2008.

Lee, T., Sullivan, A. P., Mack, L., Jimenez, J. L., Kreidenweis, S. M., Onasch, T. B., Worsnop, D. R., Malm, W. C., Wold, C. E., Hao, W. M., and Collett, J. L.: Chemical smoke marker emissions during flaming and smoldering phases of laboratory open burning of wildland fuels., Aerosol Sci. Tech., 44, 9, I–V, doi:10.1080/02786826.2010.499884, 2010.

Ludwig, J., Marufu, L. T., Huber, B., Andreae, M. O., and Helas, G.: Domestic combustion of biomass fuels in developing countries: A major source of atmospheric pollutants, J. Atmos. Chem., 44, 23–37, 2003.

McKeen, S. A., Liu, S. C., Hsie, E. Y., Lin, X., Bradshaw, J. D., Smyth, S., Gregory, G. L., and Blake, D. R.: Hydrocarbon ratios during PEM-WEST a: A model perspective, J. Geophys. Res.-Atmos., 101, 2087–2109, 1996.

Muhle, J., Lueker, T. J., Su, Y., Miller, B. R., Prather, K. A., and Weiss, R. F.: Trace gas and particulate emissions from the 2003 Southern California wildfires, J. Geophys. Res.-Atmos., 112, D03307, doi:10.1029/2006jd007350, 2007.

Narukawa, M., Kawamura, K., Takeuchi, N., and Nakajima, T.: Distribution of dicarboxylic acids and carbon isotopic compositions in aerosols from 1997 Indonesian forest fires, Geophys. Res. Lett., 26, 3101–3104, 1999.

Peltier, R. E., Hecobian, A. H., Weber, R. J., Stohl, A., Atlas, E. L., Riemer, D. D., Blake, D. R., Apel, E., Campos, T., and Karl, T.: Investigating the sources and atmospheric processing of fine particles from Asia and the Northwestern United States measured during INTEX B, Atmos. Chem. Phys., 8, 1835–1853, doi:10.5194/acp-8-1835-2008, 2008.

Penner, J. E., Charlson, R. J., Hales, J. M., Laulainen, N. S., Leifer, R., Novakov, T., Ogren, J., Radke, L. F., Schwartz, S. E., and Travis, L.: Quantifying and minimizing uncertainty of climate forcing by anthropogenic aerosols, B. Am. Meteorol. Soc., 75, 375–400, 1994.

Reddy, M. S. and Boucher, O.: A study of the global cycle of carbonaceous aerosols in the LMDZT general circulation model, J. Geophys. Res.-Atmos., 109, D14202, doi:10.1029/2003jd004048, 2004.

Reid, J. S., Eck, T. F., Christopher, S. A., Koppmann, R., Dubovik, O., Eleuterio, D. P., Holben, B. N., Reid, E. A., and Zhang, J.: A review of biomass burning emissions part III: intensive optical properties of biomass burning particles, Atmos. Chem. Phys., 5, 827–849, doi:10.5194/acp-5-827-2005, 2005.

Sachse, G. W., Hill, G. F., Wade, L. O., and Perry, M. G.: Fast-response, high-precision carbon-monoxide sensor using a tunable diode-laser absorption technique, J. Geophys. Res.-Atmos., 92, 2071–2081, 1987.

Schmidl, C., Bauer, H., Dattler, A., Hitzenberger, R., Weissenboeck, G., Marr, I. L., and Puxbaum, H.: Chemical characterization of particle emissions from burning leaves, Atmos. Environ., 42, 9070–9079, doi:10.1016/j.atmosenv.2008.09.010, 2008.

Scholes, M. and Andreae, M. O.: Biogenic and pyrogenic emissions

from Africa and their impact on the global atmosphere, Ambio, 29, 23–29, 2000.

Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics: From air pollution to climate change, John Wiley, NY, 1998.

Simpson, I. J., Akagi, S. K., Barletta, B., Blake, N. J., Choi, Y., Diskin, G. S., Fried, A., Fuelberg, H. E., Meinardi, S., Rowland, F. S., Vay, S. A., Weinheimer, A. J., Wennberg, P. O., Wiebring, P., Wisthaler, A., Yang, M., Yokelson, R. J., and Blake, D. R.: Boreal forest fire emissions in fresh Canadian smoke plumes: C1–C10 volatile organic compounds (VOCs), CO_2, CO, NO_2, NO, HCN and CH_3CN, Atmos. Chem. Phys., 11, 6445–6463, doi:10.5194/acp-11-6445-2011, 2011.

Singh, H. B., Anderson, B. E., Brune, W. H., Cai, C., Cohen, R. C., Crawford, J. H., Cubison, M. J., Czech, E. P., Emmons, L., Fuelberg, H. E., Huey, G., Jacob, D. J., Jimenez, J. L., Kaduwela, A., Kondo, Y., Mao, J., Olson, J. R., Sachse, G. W., Vay, S. A., Weinheimer, A., Wennberg, P. O., and Wisthaler, A.: Pollution influences on atmospheric composition and chemistry at high northern latitudes: boreal and California forest fire emissions, Atmos. Environ., 44, 4553–4564, 2010.

Smith, S. J., Pitcher, H., and Wigley, T. M. L.: Global and regional anthropogenic sulfur dioxide emissions, Global Planet. Change, 29, 99–119, 2001.

Stocks, B. J., Alexander, M. E., Wotton, B. M., Stefner, C. N., Flannigan, M. D., Taylor, S. W., Lavoie, N., Mason, J. A., Hartley, G. R., Maffey, M. E., Dalrymple, G. N., Blake, T. W., Cruz, M. G., and Lanoville, R. A,: Crown fire behaviour in a northern jack pine-black spruce forest, Canadian Journal of Forest Research-Revue Canadienne de Recherche Forestiere, 34, 8, 1548–1560, 2004.

Stohl, A., Forster, C., Frank, A., Seibert, P., and Wotawa, G.: Technical note: The Lagrangian particle dispersion model FLEXPART version 6.2, Atmos. Chem. Phys., 5, 2461–2474, doi:10.5194/acp-5-2461-2005, 2005.

Sullivan, A. P., Peltier, R. E., Brock, C. A., de Gouw, J. A., Holloway, J. S., Warneke, C., Wollny, A. G., and Weber, R. J.: Airborne measurements of carbonaceous aerosol soluble in water over Northeastern United States: Method development and an investigation into water-soluble organic carbon sources, J. Geophys. Res.-Atmos., 111, D23S46, doi:10.1029/2006jd007072, 2006.

Trentmann, J., Andreae, M. O., and Graf, H. F.: Chemical processes in a young biomass-burning plume, J. Geophys. Res.-Atmos., 108, 4705, doi:10.1029/2003jd003732, 2003.

Turetsky, M. R., Kane, E. S., Harden, J. W., Ottman, R. D., Maines, K. L, Hoy, E., and Kashschke, E. S.: Recent acceleration of biomass burning and carbon losses in Alaskan forests and peatlands, Nature Geosci., 4, 27–31, 2011.

van Donkelaar, A., Martin, R. V., Leaitch, W. R., Macdonald, A. M., Walker, T. W., Streets, D. G., Zhang, Q., Dunlea, E. J., Jimenez, J. L., Dibb, J. E., Huey, L. G., Weber, R., and Andreae, M. O.: Analysis of aircraft and satellite measurements from the Intercontinental Chemical Transport Experiment (INTEX-B) to quantify long-range transport of East Asian sulfur to Canada, Atmos. Chem. Phys., 8, 2999–3014, doi:10.5194/acp-8-2999-2008, 2008.

Vay, S. A., Woo, J. H., Anderson, B., Thornhill, K. L., Blake, D. R., Westberg, D. J., Kiley, C. M., Avery, M. A., Sachse, G. W., Streets, D. G., Tsutsumi, Y., and Nolf, S.: The influence of regional-scale anthropogenic emissions on CO_2 distributions over the Western North Pacific, J. Geophys. Res.-Atmos., 108, 8801, doi:10.1029/2002JD003094, 2003.

Vay, S. A., Choi, Y., Vadrevu, K. P., Blake, D. R., Tyler, S. C., Wisthaler, A., Hecobian, A., Kondo, Y., Diskin, G. S., Sachse, G. W., Woo, J.-H., Weinheimer, A. J., Burkhart, J. F., Stohl, A., and Wennberg, P. O.: Patterns of CO_2 and radiocarbon across high northern latitudes during International Polar Year 2008, J. Geophys. Res.-Atmos., 116, D14301, doi:10.1029/2011JD015643, 2011.

Warneke, C., Bahreini, R., Brioude, J., Brock, C. A., de Gouw, J. A., Fahey, D. W., Froyd, K. D., Holloway, J. S., Middlebrook, A., Miller, L., Montzka, S., Murphy, D. M., Peischl, J., Ryerson, T. B., Schwarz, J. P., Spackman, J. R., and Veres, P.: Biomass burning in Siberia and Kazakhstan as an important source for haze over the Alaskan Arctic in April 2008, L02813, Geophys. Res. Lett., 36, 2813–2813, 2836, doi:10.1029/2008GL036194, 2009.

Weinheimer, A., Walega, J., Ridley, B. A., Gary, B. L., Blake, D. R., Rowland, F. S., Sachse, G. W., Anderson, B., and Collins, D. R.: Meridional distributions of NO_x, NO_y and other species in the lower stratosphere and upper troposphere during AASE-II, Geophys. Res. Lett., 21, 2583–2586, 1994.

Wisthaler, A., Hansel, A., Dickerson, R. R., and Crutzen, P. J.: Organic trace gas measurements by PTR-MS during INDOEX 1999, J. Geophys. Res.-Atmos., 107, 8024, doi:10.1029/2001JD000576, 2002.

Yamasoe, M. A., Artaxo, P., Miguel, A. H., and Allen, A. G.: Chemical composition of aerosol particles from direct emissions of vegetation fires in the Amazon basin: Water-soluble species and trace elements, Atmos. Environ., 34, 1641–1653, 2000.

Yokelson, R. J., Christian, T. J., Karl, T. G., and Guenther, A.: The tropical forest and fire emissions experiment: laboratory fire measurements and synthesis of campaign data, Atmos. Chem. Phys., 8, 3509–3527, doi:10.5194/acp-8-3509-2008, 2008.

Yokelson, R. J., Crounse, J. D., DeCarlo, P. F., Karl, T., Urbanski, S., Atlas, E., Campos, T., Shinozuka, Y., Kapustin, V., Clarke, A. D., Weinheimer, A., Knapp, D. J., Montzka, D. D., Holloway, J., Weibring, P., Flocke, F., Zheng, W., Toohey, D., Wennberg, P. O., Wiedinmyer, C., Mauldin, L., Fried, A., Richter, D., Walega, J., Jimenez, J. L., Adachi, K., Buseck, P. R., Hall, S. R., and Shetter, R.: Emissions from biomass burning in the Yucatan, Atmos. Chem. Phys., 9, 5785–5812, doi:10.5194/acp-9-5785-2009, 2009.