



Quantifying gaseous emissions from tropical savanna fires in northern Australia using UV-DOAS and FTIR remote sensing.

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Abstract

IR and UV measurements of open vegetation fire plumes were recorded during the 2009 WALFA campaign in Arnhem Land, Australia. Managed fires were conducted in the early (June/July) and late (September/October) dry season, allowing seasonal emission comparisons. Pathlength concentrations of the main plume gaseous species (CO₂/CO/CH₄) were quantified using Fourier Transform Infrared (FTIR) Spectroscopy. Observed concentrations were subsequently used to calculate emission ratios and emissions factors for the target gases. Current literature values of emission ratios/factors are associated with large inter-fire variations/uncertainties. A UV-DOAS instrument was used to quantify NO₂ and SO₂ emissions, allowing the determination of flux-rates when coupled with plume velocity estimates. SO₂ fluxes exceeded NO₂ fluxes, suggesting possibly low nitrogen content in the study ecosystem. Assuming NO₂/SO₂ are detectable by the FTIR, DOAS flux-rates and FTIR emissions ratios can be combined to return flux-rates of all FTIR detectable species. Results thus far advocate using these two instruments in relation to trace gas measurements from vegetation fires. Derived fluxes are compared with the fire radiative power (FRP) time-series, calculated from airborne thermal imaging. Flux rates show a strong correlation with FRP, confirming the relationship between fuel FRP and fuel consumption rate at the scale of these fires.

Keywords: Biomass, DOAS, FTIR, Emissions, Plume

1. Introduction

The amount of severe fire activity over the last decade, as well as the increase in scientific interest in the area, has led to a rise in public and scientific recognition of the importance of emissions from biomass burning and, in particular, their effects on the atmosphere (Andreae and Merlet, 2001). Accurate data on biomass burning emissions are required to understand land-atmosphere interactions and the varying processes that control Earth's atmospheric composition, chemistry and climate at a variety of temporal scales (Ito and Penner 2004). Remote sensing is an important tool enabling quantification of many aspects of biomass burning, including source location, burned area, strength and timing, effects of smoke on chemistry and radiative budget, and how these vary over both space and time (e.g. Parasad *et al.* 2002, Riggan *et al.* 2004). Such data may also allow us to better model the changes that may occur in the future due to the impact of climate change on fire regimes, and also to realize strategies for the mitigation of and/or adaptation to these changes.

The WALFA (West Arnhem Land Fire Abatement Project) project is an initiative based on investigating the impacts of severe wildfires and fire management issues in the indigenous owned savanna ecosystems of western Arnhem Land. The study site is located at the upper reaches of the Liverpool River, western Arnhem Land and is frequently subject to large fires, most notably during the late dry season. One focus of the WALFA project is to incorporate the potential for GHG emissions reduction into fire management practices. Preliminary results presented here make a contribution to the WALFA 2009 study, and focus on a comparison of GHG emissions from biomass burning fires occurring in the early (July) and late (Oct) dry season of 2009.



Emissions are determined here using analysis of spectra measured via open path Fourier Transform Infrared (FTIR) spectroscopy and Ultra-Violet Differential Optical Spectroscopy (UV-DOAS) of the biomass burning smoke.

This work is part of a wider PhD project investigating trace gas emissions from biomass burning worldwide, and in particular the variation in emissions ratios (the amount of a particular gas released as a function of another reference gas [e.g. CO₂ or CO] and emissions factors (the amount of a particular gas released as a function of the amount of dry fuel burnt). Emissions factors are important in quantifying the impact of biomass burning since they are used in almost all emissions modelling calculations (e.g. van der Werf *et al.* 2006, Ito and Penner 2004). However, there is a lack of up-to-date, consistent literature on these factors, especially with respect to space, time and combustion type variations. Most current emissions ratios and factors are obtained from a mixture of laboratory and *in situ* sample analysis (Jain *et al.* 2006). This study aims to contribute to the development of more wide-ranging and robust emissions inventories, and for the WALFA2009 observations the FTIR-derived emissions ratios and emissions factors will be combined with in-plume concentrations of NO₂ and SO₂ measured using the UV-DOAS in order to estimate the flux rates of all target species (including GHGs). Flux rates are compared with simultaneous Fire Radiative Power (FRP) measures determined via airborne thermal imaging, since this is also believed to be directly related to the rate of biomass combustion (Wooster *et al.* 2005).

2. Methodology

2.1 Sampling Techniques

An IR lamp was employed as the radiation source for the FTIR spectrometer observations. FTIR and source were positioned stationary, slightly downwind from the fire, and approximately 40 m apart such that the plume was able to pass between them. IR spectral measurements were taken before, during and after the fire, with the non-fire measurements used to quantify the background concentration of each target gas. Castro *et al.* (2007) and Karl *et al.* (2007) describe similar OP-FTIR setups. The UV-DOAS instrument was positioned pointing zenith on a vehicle that traversed repeatedly back and forth alongside the plot, underneath the plume and approximately perpendicular to its direction of travel. The mini DOAS instrument deployed here has previously been used to monitor air pollution (e.g. Theys *et al.* 2007) and volcanic plumes (e.g. Galle *et al.* 2002, McGonigle 2007), and we know of one example of its use with fire smoke (Oppenheimer *et al.* 2004). DOAS spectra from UV-transparent gas cells containing known concentrations of SO₂ and NO₂ were measured before and after each fire for calibration purposes. A MIR (3.9 μm) thermal camera was operated in a helicopter hovering above the fire plot to record the fire-emitted thermal radiation for the purposes of FRP determination.

2.2 Data Analysis

Pathlength in-plume concentrations of the target gases from the FTIR spectra were retrieved using MALT (Griffith, 1996), a non-linear, least squares fitting algorithm based on forward modelling of the recorded IR spectra. The algorithm works within a given fitting wavelength region, using estimates of temperature and pressure combined with *a priori* values of the target gas concentrations to create a synthetic IR spectrum which is iteratively compared with the measured spectrum until a best match is found. Emission factors and emission ratios were derived from the retrieved in-plume pathlength trace gas concentrations using the approaches outlined by Andreae and Merlet (2001).

The empirical FLYSPEC method, as used previously by Elias *et al.* (2005) was used to retrieve plume pathlength concentrations of SO₂ and NO₂. Gas cell calibration measurements are used to determine a quadratic relationship between spectral absorption peaks and trace gas concentration. This relationship is subsequently applied to compute smoke SO₂ and NO₂ concentrations. Flux rates of SO₂ and NO₂ were then determined using the DOAS-derived trace gas column densities, a GPS-record of the vehicle traverse, and estimates of plume velocity derived from wind-speed measurements made simultaneous with the fire and adjusted for the estimated plume height using the log-law for wind. This approach has been widely adopted for the analysis of volcanic plumes SO₂ fluxes (e.g. Oppenheimer *et al.* 2004, McGonigle 2007). The derived flux rates of SO₂ and NO₂ can be converted into flux rates of GHG species via use of the GHG/SO₂ and GHG/NO₂ emissions ratios, derived from the FTIR data.

The 1 Hz thermal camera data were converted to an FRP time series using a simple temperature thresholding algorithm to detect fires pixels and then calculating the per-pixel FRP using the MIR method of Wooster *et al.* (2005). The total FRP of each image frame was calculated by summing the FRP of each fire pixel present and the resultant 1 second temporal resolution FRP time series used to compare trace gas fluxes with the fire radiant heat release rate.

4. Results

Results have so-far been derived for sample fires conducted during the early (04-07-2009) and late (01-10-09) season campaigns respectively. The early season fire was burnt in primarily grassy fuels, whilst the late season fire was mainly fueled by leaf litter. Presented in the results are the FTIR-derived in-plume gas concentrations of several species, their corresponding emission ratios and emissions factors, the in-plume concentrations of SO₂ and NO₂ retrieved from the UV-DOAS analysis, and a first comparison between trace gas flux rates and FRP.

4.1 DOAS derived flux rates.

Figure 1(A) and (B) shows the flux rates for NO₂ and SO₂ for the early and late season fires respectively, along with the matching FRP time series. Note that the fluxes have not yet been adjusted for situations when the plume and vehicle traverse directions were non-orthogonal. Figure 2 presents the in-plume pathlength concentrations of CO₂, CO and CH₄ retrieved from the FTIR-spectra for the (A and B) early and (C and D) late dry season fires. These data were subsequently used to calculate the trace gas emission ratios and emission factors for each fire.

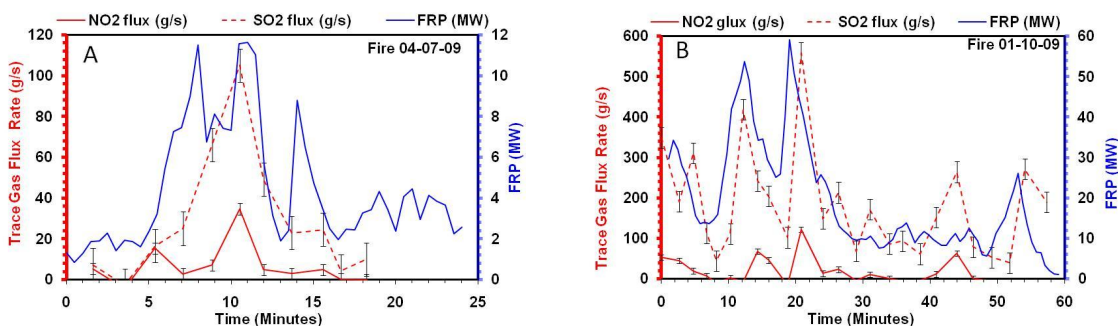


Figure 1: Flux rates of NO₂ and SO₂ derived via analysis of UV-DOAS spectra and plume velocity estimates. Also shown are the matching FRP time series determined from airborne thermal imaging. (A) early dry season and (B) late dry season fires. Note the strong relationship between the FRP time series and gas flux.

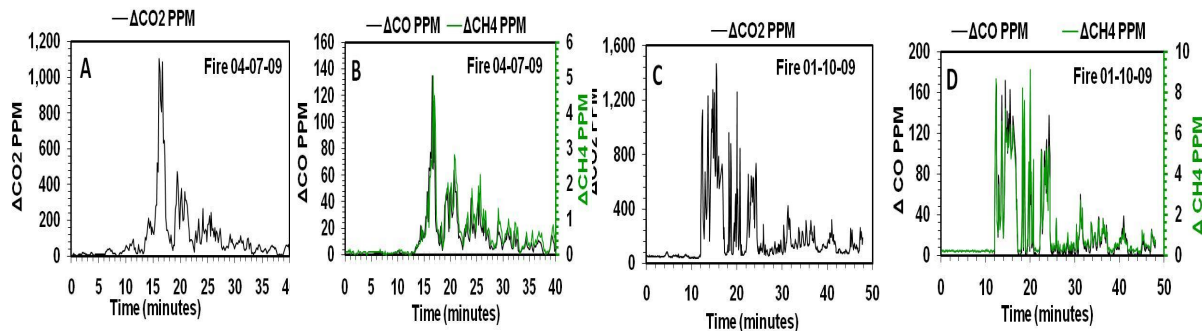


Figure 2: FTIR-derived above background pathlength concentrations of CO₂ and CO for (A, B) early dry season fires, and (C, D) late dry season fires. Note the large emission peaks reaching well above background concentrations for all species within the plume.



5. Discussion / Analysis

The FTIR-derived results indicate significant emissions of the main carbon containing trace gas species from both early and late dry season fires. Comparison of emission factors between the early and late season fires show little difference for CO₂ and CO, but for CH₄ the emission factors for grass and leaf litter fuel types is 20% - 60% higher for the late season fires than for the early season fires. Therefore, in this case, the late dry season fires preferentially released more CH₄ per unit of fuel burned than did the early dry season fires.

Emissions of SO₂ appear greater in all fires than does the emission of NO₂. This may result from a relative lack of nitrogen in the burned ecosystems, which will be investigated. Trace gas flux rates show a good match to the FRP time series, supporting the use of FRP measures in the estimation of fuel consumption and trace gas emission rates (Wooster *et al.* 2005). Flux rate calculations need further refinement, in particular the approach used to link the DOAS-derived fluxes with the FTIR measured in-plume GHG and SO₂/NO₂ concentrations. Currently, the FTIR-derived SO₂ time series has been used, however the IR absorption bands for SO₂ are weak and result in significant uncertainties. An alternative is the targeting of smoke formaldehyde (H₂CO), which has strong absorption bands in both the UV and IR. However calibration gas cells for use with the UV-DOAS instrument are unavailable for this gas, hence the use of synthetic spectra will be investigated for this purpose.

6. Conclusions

Preliminary results confirm the ability of both OP-FTIR and UV-DOAS spectroscopy in quantifying gas emissions from biomass burning plumes, both in relation to absolute plume pathlength concentrations and flux rates. Preliminary results show higher emission factors for leaf litter and grass fires for CH₄ towards the later burning season as opposed to the early dry season. The use of UV-DOAS to return in-plume flux rates from biomass burning events is little used (currently just Oppenheimer *et al.*, 2004) and the synthesis of UV-DOAS and FTIR approaches conducted here has not been previously implemented in the area of fire emissions. The ultimate aim of the project is to optimise and deploy the methodology presented here to investigate a wider range of biomass burning scenarios and provide a source of emissions ratio and emissions factor data with potential for use in the modelling of biomass burning emissions in a variety of fire affected regions.

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