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Estimated total emissions of trace gases from the Canberra wildfires of 2003: a new method using satellite measurements of aerosol optical depth and the MOZART chemical transport model

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Abstract

In this paper we describe a new method for estimating trace gas emissions from large vegetation fires using measurements of aerosol optical depth from the MODIS instruments onboard NASA's Terra and Aqua satellites, combined with the atmospheric chemical transport model MOZART. The model allows for an estimate of double counting of enhanced levels of aerosol optical depth in consecutive satellite overpasses. Using this method we infer an estimated total emission of 10±3 Tg of carbon monoxide from the Canberra fires of 2003. Emissions estimates for several other trace gases are also given. An assessment of the uncertainties in the new method is made and we show that our estimate agrees (within expected uncertainties) with estimates made using current conventional methods of multiplying together factors for the area burned, fuel load, the combustion efficiency and the emission factor for carbon monoxide. The new method for estimating emissions from large vegetation fires described in this paper has some significant uncertainties, but these are mainly quantifiable and largely independent of the uncertainties inherent in conventional techniques. Thus we conclude that the new method is a useful additional tool for characterising emissions from vegetation fires.

Introduction

The Canberra wildfires of 2003

Abnormally high temperatures and low rainfall were experienced in south eastern Australia in the second half of 2002 due to a severe El Niño related drought that created conditions of extreme fire danger, with an abundance of dry fuel ready to burn. On 8 January 2003, fires were started by lightning strikes in the region around the Australian capital city of Canberra (35° S, 149° E) and the nearby Snowy Mountains (Webb et al., 2004). The fires burned for several weeks, reaching their maximum intensity on 18

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January 2003 when a firestorm engulfed several outer suburbs of Canberra. Four people died and there was widespread damage to property including 491 houses lost. The fires also caused extensive damage to rural grazing property, forests and bushland in the area with substantial losses of farming stock and wildlife (Webb et al., 2004). At the peak of the firestorm on 18 January 2003 there were pyro-cumulonimbus eruptions that injected smoke plumes into the stratosphere (Fromm et al., 2006). The fires continued throughout January and well into February until they were eventually extinguished with the help of heavy rainfall towards the end of February 2003.

Smoke plumes from these fires passed over Wollongong (34°S, 151°E) and were measured by the remote sensing mid-infrared and ultra-violet/visible spectrometers at the University of Wollongong (Paton-Walsh et al., 2004; Rinsland et al., 2005). Total column amounts of trace gases derived from these mid-infrared spectra showed very strong correlations with coincident and co-located measurements of aerosol optical depth at 500 nm (Paton-Walsh et al., 2005, 2008). These strong correlations mean that it is possible to infer the enhanced column amount of a trace gas such as carbon monoxide (CO) that is present in an active fire region (whilst the smoke is still relatively young) using satellite measurements of enhanced aerosol optical depth (AOD) over the region. Combining the inferred enhancements of CO for each day that the fires burned with modelling of the dispersion of the plumes forms the basis of a new method for estimating total emissions to the atmosphere from a major fire episode. The use of aerosol optical depth to infer the emissions of trace gases such as CO introduces additional uncertainties compared to studies that directly use satellite measurements of CO, e.g. (Pfister et al., 2005), however aerosol optical depth measurements typically have greater sensitivity to the boundary layer and thus may capture smoke that could be missed by satellite-based measurements of CO. Possibly a more significant advantage of using AOD as a proxy for CO in this method is the fact that aerosols are comparatively short-lived and so provide a better indication of fresh smoke emissions than the presence of enhanced amounts of relatively long lived trace gases such as CO.

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1.2 MODIS measurements of AOD

The Moderate Resolution Imaging Spectroradiometer, (MODIS) instruments provide high radiometric sensitivity in 36 spectral bands ranging in wavelength from 0.4 µm to 14.4 µm (Kaufman, 1993; Kaufman et al., 2003, 1990; King et al., 1999; Remer et al., 2002; Zhao et al., 2003). They are carried on both the Terra and Aqua satellites (orbiting at an altitude of 705 km), providing global coverage every 1 to 2 days with a swath that is 2330-km cross track by 10 km along the track at nadir. For most parts of the globe, including south east Australia, twice daily coverage at local times of about 10:45 LT (Terra) and 14:00 LT (Aqua) is achieved, being equivalent to 00:45 UT (Terra) and 04:00 UT (Aqua). Each instrument contains a two-mirror off-axis collimating telescope, which collects radiant energy and directs it into four refractive objective assemblies, covering the visible, near infrared, short-wave to medium-wave infrared and long-wave infrared regions of the spectrum.

The MODIS instruments' products include visible images, aerosol optical depth (also called aerosol optical thickness) and thermal anomalies (pinpointing the locations of fires) (Kaufman et al., 1998). MODIS aerosol optical depth (product MOD 04) data are retrieved over the oceans globally and over large portions of the continents at the spatial resolution of 10×10 km (Chu et al., 1998; Kaufman et al., 1997; Tanre et al., 1997). Separate algorithms are used to derive aerosol optical depth over sea and land, with recent work to optimise these algorithms including the deep-blue algorithm developed to derive aerosol optical thickness over bright land areas (Levy et al., 2007; Kleidman et al., 2005).

2 Determining the increased atmospheric loading of carbon monoxide from the fires using MODIS aerosol optical depth measurements

The enhanced column amounts of carbon monoxide that were present in the atmosphere in the region surrounding the Canberra fires of 2003 were inferred using the

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following steps:

Step 1: Average MODIS AOD data

MODIS product "MOD 04" aerosol optical depth measurements over Southeastern Australia from 7 January to 26 February 2003 were re-projected and plotted as a series of maps; one for each satellite overpass, giving two images per day. Many of the images have large amounts of missing data where the algorithms have rejected the data either as a result of cloud interference, sea-glint or other technical difficulties, resulting in interruptions in the twice daily time-series of images. When one of the two images for the day showed a significantly clearer view of plumes from the fires, then only this image was used. Otherwise data from both images were combined. All aerosol optical depth values from the chosen images were averaged over 1°×1° grid boxes, extending from -25° to -46° latitude and 135° to 160° longitude. Example images are shown in Fig. 1.

Step 2: Determine the background AOD before the fires began

The 1° by 1° averaged AOD data from the Aqua overpass from 7 January 2003 (the day before the fires started) were used to establish the background aerosol optical depth in the fire-affected area. There are sources of enhanced AOD other than smoke and so the background must be chosen carefully so as not to overestimate the emissions by including AOD that results from dust or sea salt aerosols. Only 1° by 1° grid boxes with more that 10 points were considered because of the possibility of getting a few spuriously high points adjacent to points that are rejected by the MODIS algorithm due to cloud interference. The maximum AOD value for a 1° by 1° grid box on 7 January before the fires started was 0.2, and so this was taken as a threshold value to identify smoke. The average AOD value for a 1° by 1° grid box on 7 January before the fires started was 0.11 and this was used as a "background" value. In the subsequent days, whilst the fires burned, AOD values above 0.2 are assumed to result from particulates in the smoke emitted from the fires. Each grid box with an averaged AOD value above 0.2 was included in the calculation and the background value of 0.11 subtracted to yield the enhancements in AOD assumed to result from the smoke plumes.

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(AOD_{excess}).

Step 3: Convert enhanced AOD to enhanced mass of carbon monoxide

The 1° by 1° averaged AOD excess values were converted to equivalent enhanced carbon monoxide (CO) column amounts (molecules cm⁻²) via the relationship CO=(1.5±0.1)AOD×10¹⁸ molecules cm⁻², as determined in Paton-Walsh et al. (2005). The enhanced CO column amounts were then converted to enhanced atmospheric loading of CO (g) by multiplying by the area of the 1° by 1° grid boxes (in square centimetres) and the molecular weight of CO and dividing by Avogadro's number.

The conversion is given by Eq. (1) below:

$$M_{\rm CO} = \frac{{\rm AOD}_{\rm excess} G_{\rm CO:AOD} A_{\rm grid}}{N_{\Delta}} {\rm MW}_{\rm CO}$$
 (1)

Where

- $M_{\rm CO}$ =enhanced mass of CO in the region as a result of the fires
- AOD_{excess} is 0.11 less than the 1° by 1° grid averaged MODIS AOD in each grid box that had a value above 0.2
- $G_{\text{CO:AOD}}$ is 1.5×10¹⁸ molecules cm⁻² (the gradient of CO to AOD determined in Paton-Walsh et al., 2005)
- $-A_{grid}$ is 1.02×10^{14} the area of the grid boxes in cm² using conversion factors of 111.12 km for 1° latitude and 92.12 km for 1° longitude
- N_A is Avogadro's number= 6.02×10^{23} mol⁻¹ and
- MW_{CO} is the molecular weight of CO (28 g mol⁻¹)

Finally summing the mass of enhanced CO in all the contributing grid boxes gives the total enhanced atmospheric mass of CO in the area on each day, (50 days from 8 January to 26 February 2003). Note that the enhanced mass of any trace gas that is present in any large active fire region (for which there is a known strong correlation to AOD in young smoke plumes) may be inferred using the method outlined above.

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A new method for estimating emissions using MODIS aerosol optical depth measurements

For every day from 8 January to 26 February 2003 the MODIS AOD measurements have yielded a daily value for the enhanced mass of CO over the defined fire region. A simple addition of these daily enhancements of CO yields a total amount of 12 teragrams (Tg) of CO. However smoke that remains in the defined region of interest for more than 24 h after being emitted by the fires has the potential to be detected by the MODIS AOD measurements from more than one satellite overpass. Thus the simple addition of daily enhancements in mass of CO may yield an overestimate of the emissions from the fires if double counting of non-dispersed smoke is more significant than smoke that goes undetected (either because it is transported out of the sampled region before the next satellite overpass or because of missing MODIS data in the daily images). For this reason it is necessary to model the dispersion of the plumes.

3.1 Modelling the dispersion of the smoke plumes

An estimate of the CO that remained in the atmosphere from emissions from fires on previous days was made using two separate model runs. The model chosen was the offline global chemical transport model MOZART-4 (Model of Ozone And Related Tracers-Version 4) (Emmons et al., 2009). The model was driven by meteorology from the NCAR reanalysis of the National Centers for Environmental Prediction (NCEP) forecasts (Kalnay et al., 1996) at a horizontal resolution of approximately 2.8° by 2.8°, with 28 vertical levels from the surface to approximately 2 hPa. An initial model run incorporated full chemistry and all the standard emission inventories used in MOZART-4 (except that 8-day averages for biomass burning from van der Werf et al. (2006) were used in place of the usual monthly averages from the same source). The concentration fields for OH were extracted from this first run and used to define the atmospheric concentrations of OH for chemistry within the second run. In reality chemical removal of CO by reaction with OH will have very little effect on the CO concentrations in the

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region of the fires compared to the effects of smoke dispersion. If this were not the case then the CO chemistry would need to be omitted in the second model run, since the measurements actually depend upon the aerosol properties within the smoke plumes not the CO.

The second simulation was a simple CO tracer simulation using the archived OH distributions from the first run. The only emissions released in the model were CO emissions from the Canberra fires, with a separate tagged CO emission source defined for each day of the fires from 8 January to 26 February 2003. The locations of the fires were estimated from images of MODIS fire scars and thermal hotspots and then used to define the area over which the emissions were released into the model each day. In total seven 0.5° by 0.5° grid boxes were used as emission sources centred on the following coordinates: (35.5° S, 149.0° E), (36.0° S, 148.5° E), (36.5° S, 148.5° E), (37.0° S, 148.5° E), (37.0° S, 147.0° E). The number of these grid boxes actually used as the source area for each day's emissions varied from only four to all seven, depending upon the location of the thermal hotspots detected that day. The flux rate for each tagged source was defined by releasing the inferred total mass of enhanced CO from the relevant day from the chosen source area at a uniform rate over the preceding 24 h.

The second model run started on 1 January 2003 and continued until 26 February with 48 individually tagged days of CO emissions. MOZART models the dispersion of the emitted CO and the mass of each tagged CO source that remains in the defined region (from -25° to -46° latitude and 135° to 160° longitude) is extracted at 6-hly intervals from the modelled concentration fields.

3.2 Using the model output to correct for double-counting of the smoke

The inferred enhancement of atmospheric mass of CO from the second day of the fires is adjusted to account for smoke emitted on the first day that MOZART predicts will still be in the defined region a day later. Not all of the smoke remaining in the region will be captured by the MODIS AOD measurements because of the number of grid boxes

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with missing data. The estimated double counting of CO on day 2 is the modelled remaining mass of tagged CO from day 1 multiplied by the fraction of grid boxes with real MODIS AOD values (not missing data). The estimated amount of double counting is then subtracted from the enhanced CO in the region to yield the estimated fresh emissions during the second day of the fires.

Day 3 emissions must be adjusted for smoke remaining from day 1 and day 2 of the fires. However the tagged emissions released into the model on day 2 of the fires were the total enhanced mass of CO in the region inferred from the satellite AOD image via Eq. (1) before adjusting for double counting. Thus the mass of tagged emissions remaining in the model from day 2 must be decreased by the ratio:

before summing with the day 1 emissions still remaining. This sum is then multiplied by a factor to account for missing data to yield an estimate of the double-counting on day 3.

Note that the calculation of emissions from later days in the fires requires the estimates from previous days. Thus the emissions must be calculated day by day, each time adjusting the emissions in the model to account for double-counting of smoke emitted on previous days.

The process may be generalised by Eqs. (2) and (3):

$$E_{\text{CO-day}_{a}} = M_{\text{CO-day}_{a}} - DC_{\text{CO-day}_{a}}$$
 (3)

where

- $E_{\text{CO-day}_n}$ = estimated mass of CO emitted by the fires during day n
- $M_{\text{CO-day}_n}$ = enhanced mass of CO in the region after n days of fires, inferred from the MODIS AOD measurements via Eq. (1)
- DC_{CO-day_n} = estimated double counting of smoke on day n

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and

$$DC_{CO-day_n} = \sum_{i=x}^{i=1} \left(\frac{E_{CO-day_{n-i}}}{M_{CO-day_{n-i}}} MR_{CO_tag(n-i)_day_n} \right) \cdot F_{NWPn}$$
(4)

where

- $MR_{CO_tag(n-i)_day_n}$ = the tagged mass of CO emitted on day n-i that MOZART predicts still remains in the region at the time of the satellite overpass after day n
- $-F_{NWPn}$ =fraction of real AOD values (not missing data) in the MODIS image of smoke plumes after day n of the fires

Note that in Eq. (3) adjustment is made for "x" days of double-counting. The guestion of how many days of previous emissions should be accounted for is not a simple one as it depends upon the atmospheric lifetime of the emitted aerosols. (Remember that the enhancement in CO from the fires is calculated from MODIS measurements of AOD). The lifetime of aerosols in the atmosphere varies from seconds to days depending upon the size of the particles and the altitude that they are located, with lifetimes increasing for larger particles and higher altitudes (Williams et al., 2002). The AOD of smoke around Canberra during the fires was studied using sun-photometers (Mitchell et al., 2006), however it is not clear whether or not there was a significant component of fresh smoke contributing to the AOD from fires still burning in the area. Thus the changing particle size distributions and plume heights for all the different smoke plumes emitted are not known. Instead, evidence is provided by satellite-based measurements of AOD made after the intense firestorms that burned across the Australian state of Victoria on 7 February 2009. AOD data from NASA's Ozone Monitoring Instrument (OMI) show that smoke from these intense fires separated from subsequent plumes and was transported to the north of New Zealand where it persisted for over a week before moving west across Northern Australia and dissipating entirely over the Indian Ocean by early March (Paton-Walsh et al., 2009). This shows that smoke plumes from

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intense burning of eucalyptus trees (similar to the Canberra fires of 2003) produces plumes in which significant enhancements in AOD may persist for more than a week.

Global model estimates for a lifetime of carbon aerosol of just under 5 days have been made by Rasch et al. (2000) and Chin et al. (2002), whilst Edwards et al. (2006a) use MODIS AOD and MOPITT CO measurements from the Terra satellite (Edwards et al., 2006b) to estimate a lifetime of 3.8±0.8 days. Clearly it is usual for some proportion of smoke aerosols to persist for two or three weeks as plumes may be tracked by satellite measurements over such timescales e.g. (Radhi et al., 2009; Singh et al., 2006; Paton-Walsh et al., 2009).

Given all of the available evidence we decided to correct for double-counting of smoke over 3 days. Although some proportion of the enhanced AOD probably persists for significantly longer than this our correction for double counting does not model the decay of the AOD but simply counts it as still present (at 100%) or not present (0%). In order to test the sensitivity of the resulting emissions estimates to the assumptions made for the AOD lifetime, we provide three separate estimates for the total mass of emitted CO that account for 7 days, 3 days and 1 day of double-counting of smoke.

As expected, the amount of double-counting predicted by the model is very variable from day to day as the meteorological conditions change. However the total amount of detected enhanced AOD that the model predicts results from double-counting of previous emissions over the entire 50 day period is 26% for the scenario that the AOD persists for 1 day, 44% for the 3 day scenario and 52% for the 7 day scenario.

4 Uncertainties and biases in the method for estimating total emissions from fires

Clearly there are a number of significant uncertainties in the method described here for estimating the total emissions of carbon monoxide (or other trace gases) from large-scale fires. There are significant uncertainties that pertain to correctly assigning the excess AOD that results from the fires and the degree of double-counting that occurs,

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but there are also uncertainties in the conversion from excess AOD to excess carbon monoxide. Many of the uncertainties inherent in this method are difficult to quantify but the most significant sources of uncertainty are described below along with an estimate of their probable magnitude.

5 4.1 Uncertainties in determining the enhancements in AOD at 500 nm from the fires

The major uncertainties in assigning the excess AOD from the fires come from undetected smoke and the choices of appropriate background AOD values. Undetected enhancements in AOD can result from smoke missed between clear satellite images or lost in the grid boxes with missing MODIS data leading to an underestimate of the emissions. If we make the assumption that the missing data are equally likely to contain smoke as the grid boxes with real MODIS AOD values, then we can estimate the magnitude of this uncertainty. The average percentage of grid boxes with missing data in the chosen daily images of the region around the Canberra fires is 34%. If the undetected smoke is not dispersed from the region then it may be detected the following day so the missing data count must be adjusted by the amount that is modeled as being lost from the region each day. MOZART predicts that an average of 50% of emitted CO remains in the region a day later, so the missing data count is multiplied by the 50% of smoke that will definitely not be detected the following day. This gives a negative bias of 17% from smoke missed due to grid boxes with missing data, and we can make a guess as to the uncertainty in this number by assigning half this value, i.e. ±9%.

In reality the assumption that the missing data are equally likely to contain smoke enhanced AOD as the grid boxes with real AOD values may not be true, since the images were chosen from the Terra or Aqua data (or a combination of both) not randomly but depending upon which showed the clearest image of the smoke plumes. In addition intense firestorms (like the one that occurred on 18 January 2003 near Canberra) can create pyro-cumulonimbus clouds that interfere with the satellite-instrument's ability to detect the smoke clearly, resulting in missing data as the satellite's algorithm rejects

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the data. This effect is illustrated in Fig. 2 as significant amounts of smoke are visible to the eye in the MODIS true colour image (left-hand panel) but are missed by the MODIS AOD algorithm (right-hand panel). The amount of smoke missed in this manner is not easily quantifiable but unlikely to be a major factor in the uncertainty budget since these firestorms are unusual occurrences and the smoke may yet be detected in a subsequent satellite overpass. Future improvements in the algorithms for determining the AOD of the atmosphere from satellite-based instruments like MODIS will reduce the fraction of missing data and improve the accuracy of this method.

The choice of the threshold and background values of AOD will also contribute to the uncertainties. The sensitivity of the estimated emissions to the choice of background value was estimated by using an AOD value of 0.2 for both the threshold value and the background value, which yielded a lower total emissions estimate by 20%. A value of half this difference ($\pm 10\%$) is used as an estimated uncertainty that derives from the choice of background AOD value – an uncertainty that could be either positive or negative.

4.2 Uncertainties in determining the amount of double-counting of smoke

Errors in the MOZART modeling of the emission and dispersion of smoke from the fires may lead either to double counting of smoke not truly dispersed from the region, or to wrongly identifying fresh smoke as being from earlier emissions. Hence uncertainties in the dispersion modeling can lead to either underestimates or overestimates of emissions. In evaluation studies MOZART-4 modeled output has been shown to agree well with several independent sets of observations of tropospheric chemical composition (Emmons et al., 2009) but the modeling of the emissions from the Canberra fires in the model contains significant approximations and uncertainties. In particular the true fluxes of CO from individually detected fires is not known and is approximated in the model by a uniform emission at a constant flux rate from a small number of 0.5° by 0.5° grid boxes. These approximations will lead to uncertainties in the determination of the amount of smoke remaining to be double-counted. The adjustment of the modelled

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smoke remaining by the fraction of real data points will also introduce some uncertainty since the missing data may not be equally likely to contain smoke as the data with real AOD values. Probably the largest uncertainty in the estimated double-counting comes from having to estimate the persistence of AOD within the smoke plumes from the fires. These uncertainties are extremely difficult to quantify but we can make an informed guess by comparing the double-counting predictions of 26%, 44% and 52% for the three scenarios that account for 1 day, 3 days and 7 days of double-counting, respectively. Clearly the inclusion of more and more days of potential double counting has a diminishing effect. We chose to correct for double-counting of smoke over 3 days (thereby making a reduction in the overall estimate of emissions of 44%) and assigned an estimated uncertainty of $\pm 10\%$ to this figure.

4.3 Uncertainties in converting from excess AOD at 500 nm to excess carbon monoxide

There is an uncertainty in the relationship established between ground-based sunphotometer measurements of AOD and column amounts of CO (of $\pm 7\%$) as measured by Paton-Walsh et al. (2005). In addition there are potential biases between ground-based sun-photometer AOD measurements and the 1° by 1° averaged AOD values from the MODIS instrument used in the conversion here. The viewing geometry of the two instruments differs and the satellite measurements are integrated over a large area at a single time whereas the ground-based measurements sample less than 1 m square but are averaged over several hours, therefore the values are only expected to be broadly comparable. There are only a few instances of highly elevated AOD from smoke plumes for which there are coincidental ground-based and satellite-based measurements from which a comparison may be made. Column CO amounts inferred from the MODIS AOD measurements using the nine closest 1° by 1° grid boxes to Wollongong compared to CO column amounts directly measured by ground-based Fourier transform spectrometry give a difference of 17% \pm 18%. Direct comparison between ground-based AOD measurements from Wollongong and MODIS AOD for both the

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single 1° by 1° grid box over Wollongong and the 9 closest 1° by 1° grid boxes around Wollongong, indicates that the MODIS data are on average 27%±23% lower than the equivalent ground-based data. This could be caused by instrumental, analysis, local topographic or orographic differences, leading to a potential underestimate of the emissions. Note that the column CO agreement is better because of the effect of adding in the background column of 1.5×10¹⁸ molecules cm⁻² of CO, but that the actual AOD comparison is the most relevant to the calculation of uncertainties in the overall emissions estimate.

4.4 Estimated total uncertainties in the method

The uncertainties and biases in the method for using MODIS AOD to estimate total emissions of CO from the Canberra fires are summarised in Table 1. The final emissions estimate is corrected for all of the quantified biases, although the double-counting of smoke is done on a day by day basis whilst the other corrections are applied to the end result. Since there are no obvious correlations between the different components of the uncertainty budget our best method of estimating the total uncertainty is to add all component uncertainties in quadrature, yielding a value of $\pm 30\%$. Note that the uncertainties in the conversion of sun-photometer measurements of AOD to column amounts of trace gases as measured by Paton-Walsh et al. (2005) vary from $\pm 3\%$ to $\pm 8\%$ with the result that this component makes a very small contribution to the total uncertainty. This means that the combined total uncertainties in emissions from the fires of other gases with a known correlation with AOD will also be approximately $\pm 30\%$.

In summary, there some significant uncertainties and biases in the method described in this paper, but in most part the magnitude may be estimated and the biases corrected for. Although the estimate of the overall uncertainty is $\pm 30\%$, current conventional methods of estimating emissions (Ito and Penner, 2004; Kasischke and Penner, 2004; Schultz et al., 2008; van der Werf et al., 2006) also suffer from large uncertainties, e.g., Schultz et al. quote an uncertainty in global direct carbon emissions for the 1980s and 1990s of $\pm 50\%$ and for global CO emissions for the same time period of a factor

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of 2. Importantly, the uncertainties inherent in this study are mainly independent of the uncertainties in conventional methods of estimating emissions and so this new technique is a valuable additional tool for characterising emissions from large scale vegetation fires.

Results of emissions estimate for the Canberra fires of 2003 and comparison to other inventories

As mentioned above, a simple addition of the daily enhancements of CO inferred from MODIS AOD over the defined region around Canberra (-25° to -46° latitude and 135° to 160° longitude) from 8 January to 26 February 2003 yields a total amount of 12 Tg of CO. This figure does not include any correction for double-counting, missing data or bias in the satellite AOD to sun-photometer AOD conversion. Correcting with MOZART predictions for double-counting of smoke plumes over a 3-day period and applying the other corrections reduces the value to 10.2 Tg of CO. Consideration of the total combined uncertainties gave us an estimate of ±30% and so our final estimate for the emissions of CO from the Canberra fires of 2003 is 10±3 Tg of CO.

Figure 3 is a bar chart showing each day's CO emissions alongside the modelled double counting for the day. Summing these two amounts gives the enhanced CO in the region from the fires derived from MODIS AOD data via Eq. (1). Also shown (on the right-hand axis) are the total accumulated emissions of CO from the fires.

The emissions predicted by the 8-day average GFED2 inventory (van der Werf et al., 2006) for the region from 25° to -46° latitude and 135° to 160° longitude from 8 January-26 February 2003 were extracted from the initial MOZART run described above, and gave a total emissions estimate of 13.0 Tg CO. It is possible that this emissions estimate could include other fires at the edge of the region chosen to encompass the smoke plumes from the Canberra fires. For this reason the 8-day average GFED2 inventory emissions were also extracted just for the seven 0.5° by 0.5° grid boxes that were used as emissions sources in the MOZART modelling. This gave an estimated

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total emission of 12.2 Tg CO from these fires. Van der Werf et al. (2006) do not specify a total uncertainty for the GFED2 emissions inventory but mention a figure of ±20% for burned area and must also account for uncertainties in fuel loads and emission factors (which are typically in the order of 20%–30%, Andreae and Merlet, 2001). Thus our estimate of 10±3 Tg of CO agrees with the estimate from the GFED2 inventory within the expected uncertainties.

This is a very significant emission from a single fire episode given that the mean global annual emission of CO from all vegetation fires is estimated as 330 Tg CO yr⁻¹ for the 41 years from 1960–2000 (Schultz et al., 2008) and in a separate study as 496 Tg CO yr⁻¹ for the year 2000 (Ito and Penner, 2004). Estimates for the total annual CO emissions from Australian vegetation fires in the literature include:

- 1. between 11–40 Tg CO yr⁻¹ for the years 1960–2000, averaging 23 Tg CO yr⁻¹ (Schultz et al., 2008)
- 2. 17 Tg CO yr⁻¹ for 1991 (Hurst et al., 1994)
- 3. 7 Tg CO yr⁻¹ for 2000 (Hoelzemann et al., 2004)

Thus our estimate of CO emitted from the Canberra fires actually exceeds the lowest of the estimates for the total annual CO emissions from all Australian fires for the year 2000 (Hoelzemann et al., 2004). A better comparison is the study by Shultz et al. (2008), because it covers 41 years from 1960–2000. Our estimate for the Canberra fires is approximately 45% of the average estimated annual CO emission by Shultz et al. (2008) from all Australian vegetation fires, which also includes burning of savanna grasslands and woodlands in the tropical north of Australia.

Table 2 shows the estimated total emissions from the Canberra fires for all of the trace gases measured at Wollongong by FTIR remote sensing through the smoke plumes (Paton-Walsh et al., 2005, 2008). Where available, the estimated average global annual emissions of the trace gases from all vegetation fires from Shultz et al. (2008) are also given, along with the percentage of the average global annual emissions that were emitted by the Canberra fires. By this calculation the total formaldehyde

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released by the Canberra fires represents 4.4% of the average annual global emission of formaldehyde by vegetation fires. In contrast the ethane released represents only 1% of the average annual global emissions from vegetation fires.

6 Summary and conclusions

This paper describes a new technique for estimating trace gas emissions from large vegetation fires. The new method is applied to the Canberra fires of 2003 (from 8 January to 26 February 2003), producing an emissions estimates of 10±3 Tg of carbon monoxide, which is in agreement with the emissions from the GFED2 inventory (van der Werf et al., 2006). In addition the method produces emissions estimates of 0.04±0.01 Tg of hydrogen cyanide, 0.08±0.02 Tg of ammonia, 0.17±0.05 Tg of formaldehyde, 0.024±0.007 of acetylene, 0.07±0.02 Tg of ethylene, 0.023±0.007 Tg of ethane, 0.14±0.04 Tg of formic acid and 0.20±0.06 Tg of methanol released to the atmosphere from these fires.

The dominant uncertainty in the new method arises from converting from enhanced amounts of aerosol optical depth to enhancements in trace gases. Despite this the use of aerosol optical depth has some benefits over direct satellite measurements of carbon monoxide as it provides greater sensitivity to the boundary layer and is shorter-lived and therefore potentially a better indicator of fresh smoke. There are significant uncertainties in this new technique for estimating trace gas emissions from large vegetation fires, with resulting uncertainties of $\pm 30\%$. However the uncertainties in this method are largely independent of the uncertainties in conventional methods of estimating emissions from fires, and so we conclude that this new technique is a valuable additional tool for characterising emissions from vegetation fires.

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Science Foundation. Thanks are also due to the MODIS team for providing public access to AOD data, visible images, burn scars and thermal hotspot data.

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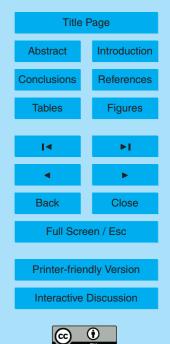


Table 1. Biases and uncertainties in the method for estimating emissions of CO from the Canberra fires.

Component of uncertainty	Applied correction to correct for bias	Uncertainty
MODIS missing data	+17%	±9%
Background AOD	n/a	±10%
Double-counting of smoke	(mean daily adjustment=-44%)	±10%
Satellite AOD to sun-photometer AOD conversion	+27%	±23%
Sun-photometer AOD to	n/a	±7%
CO conversion		
Combined total uncertainty (added in quadrature)		

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Table 2. Estimated total emissions from this study for the Canberra fires for all of the measured trace gases are shown in Tg and expressed as a percentage of the average global emissions from vegetation fires given in Schultz et al. (2008) for years 1960–1999.

Gas	Estimate of mass emitted from Canberra fires (Tg)	Average global annual emissions (Tg) Schultz et al. (2008)	% of average global annual emissions from Canberra fires
СО	10±3	330	3.1%
HCN	0.04 ± 0.01	not available	
NH_3	0.08 ± 0.02	4.90	1.5%
H₂ČO	0.17±0.05	3.90	4.4%
C_2H_2	0.024±0.007	not available	
C_2H_4	0.07 ± 0.02	4.80	1.4%
C_2H_6	0.023±0.007	2.30	1.0%
HCOOH	0.14 ± 0.04	not available	
CH ₃ OH	0.20±0.06	7.70	2.6%

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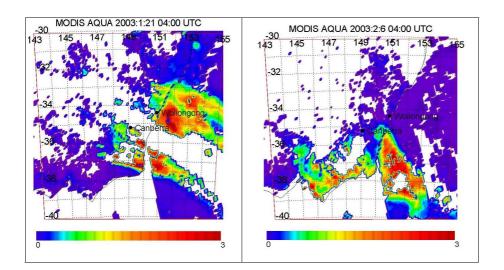


Fig. 1. Example images of MODIS product "MOD 04" aerosol optical depth measurements over Southeastern Australia re-projected and plotted as maps. The Aqua overpass from 21 January 2003 is shown in the left-hand panel and the Aqua overpass from 6 February is shown in the right-hand panel.

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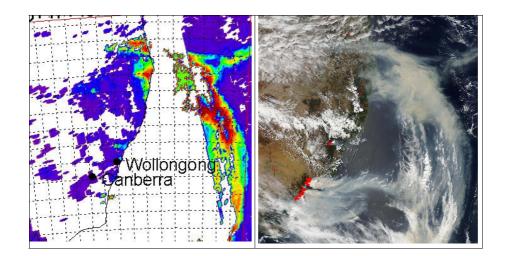


Fig. 2. Smoke plumes from the Canberra fires as seen from MODIS onboard the Aqua satellite on 18 January 2003. Left hand panel: MODIS Aerosol Optical Depth, Right-hand panel: (taken from http://rapidfire.sci.gsfc.nasa.gov/subsets/) MODIS true colour image with thermal anomalies shown as red pixels. Note that significant amounts of smoke visible to the eye in the right-hand panel are missed by the MODIS AOD algorithm.

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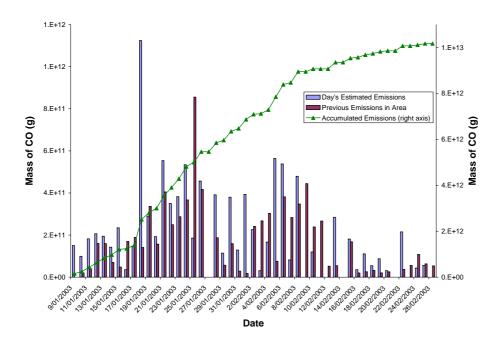


Fig. 3. The estimated emissions of CO for each day are shown alongside the previous 3 days' emissions that still remain in the area as predicted by MOZART. The accumulated total mass of CO emitted by the fires is also shown on a separate right-hand axis.

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