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Atmospheric Chemistry and Physics Discussions

Interactive comment on "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" *by* C. Paton-Walsh et al.

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The authors would like to thank both the anonymous referees for their help in improving this paper. We include below details of how we have incorporated the suggested changes into the revised manuscript. (The referee comments have been reiterated with our response below each point).

Detailed Response to Anonymous Referee #1

General Comments: (A) The paper presents a method for determining emissions of trace gases from wildfires that is complimentary to inventory and burn-scar based methods. The approach relies on an observed correlation between column CO and AOD, which it should be noted must only hold under certain conditions such as those described here (so the method may not be very general). . . .While I appreciate the effort and the goal of this work, in a way it's hard to really appreciate how useful this method is. "It almost certainly only works in cases where there aren't other sources of aerosol in the affected region (i.e., how do you screen out anthropogenic aerosols or dust, etc., if they are present?).

(1.A) This method may only be applied where measurements of AOD in relatively fresh smoke plumes (aged less than a couple of days), near major vegetation fires are available. Thus, unlike inventories based on burn-scars there is only a limited window of opportunity for the measurements to be made. It is also true that there is the potential for interfering signals from other types of aerosols like anthropogenic aerosols and dust. However it is important to note that it is only when there is a significant increase in the amount of these other pollutants that happens to coincide with the fires that there would be a problematic interfering signal. This is because only AOD values that are significantly above the normal levels for the region are considered. This is done by applying a threshold value (chosen from the maximum AOD in the region before the major fires started). So normal levels of anthropogenic aerosols from a city are not a problem unless there are suddenly enhanced well above normal levels at the same time as the fires. Similarly normal levels of atmospheric dust do not pose a problem but a dust storm that coincided with fires would be problematic.

This point has been clarified in the revised paper by adding the following:

"Note that this method is not applicable more generally to estimates of global emissions because it requires measurements of the large enhancements of aerosol optical depth that are associated with fresh smoke plumes (aged less than a couple of days) from large fire episodes. This ensures that the correlation with emitted trace gases is not compromised by aging of the plume and that (in the absence of another coincident major pollution event like a dust storm) the underlying variability in AOD from other aerosol sources will be too low to compromise the technique."

A more thorough description of the screening out of normal levels of other aerosols has been added in the method section including the following: "There are sources of enhanced AOD other than fresh smoke from the active fires and so we must be careful not to overestimate the emissions by including AOD that results from dust, sea salt aerosols or aged smoke from other fires. For this reason we impose a threshold value that is set at the maximum AOD measured in the region before the start of the large vegetation fires. By excluding all grid boxes with AOD values no higher than the maximum measured before the fires, we hope to screen out all aerosol sources other than the fires."

(B) "The authors claim uncertainty in their estimates is similar to inventory based estimates such as those from GFED." "And the uncertainty analysis seems kind of sketchy to me (i.e., "we can make a guess [that the uncertainty is] half this value [the bias due to missing grid boxes]"); a lot of the uncertainty analysis seems like guesswork. "

(1.B) Both referees raise the issue of guesswork in the uncertainty budget. This is a valid point - for many components of the uncertainty budget there is no real method of making a proper quantitative measure of the uncertainty. However, this is also true for inventory based estimates such as GFED, where there is usually a qualitative discussion of contributing uncertainties but no actual final uncertainty estimate provided: (see eg Randerson, J. T., G. R. van der Werf, L. Giglio, G. J. Collatz, and P. S. Kasibhatla. 2007. Global Fire Emissions Database, Version 2 (GFEDv2.1). Data set. Available on-line [http://daac.ornl.gov/] from Oak Ridge National Laboratory Distributed Active Archive Center, Oak Ridge, Tennessee, U.S.A.) We believe that despite its deficiencies, the uncertainty budget provided in this paper serves a useful purpose. By making an educated guess at the probable level of uncertainty in each component of the calculation - it is possible to determine how it propagates into the final uncertainty in total

emissions and thereby determine which components are likely to dominate the uncertainty budget.

Clearly this point needs to be articulated better in the paper and so we have added the following statement in the introductory paragraph to the section on uncertainties: "Many of the uncertainties inherent in this method are difficult to quantify because we do not have detailed knowledge of the true variance of the parameters. Despite this fact the exercise of constructing a full uncertainty budget can serve a useful purpose. Where there is no data on which to formally quantify the uncertainties we can make an educated guess that at the probable level of uncertainty in each component will not exceed half its value in the calculation. By this means it is possible to determine how the uncertainty propagates into the final uncertainty in total emissions. This allows us to make a very approximate estimate of the total uncertainties but also it can elucidate which components are likely to dominate the uncertainty budget."

(C) I also am skeptical, as detailed more below, about the double-counting estimate in the emissions. I think that element can particularly be addressed more reasonably the paper could be suitable for publication because it is a fairly novel idea. This has now been addressed as suggested (see point 3 below).

Specific Comments / Corrections:

1. Page 981: "MOD04" is the generic product name of the MODIS aerosol retrieval data set. But what wavelength of AOD are you using in this analysis? Presumably 550 nm, but you should say.

Yes – 550nm – corrected in text of revised paper.

2. Page 983: Regarding the model runs, the argument is that CO destruction by OH is relatively insignificant compared to dispersion of CO. This seems reasonable over the scale being considered. But I don't understand the statement that if this were not the case you should omit CO chemistry in the second model run. Why? What

measurements are you referring to in this statement?

The model was run with CO chemistry as this is the default. In reality this makes little difference as the OH chemistry is insignificant on the timescales of interest. In fact the choice of CO is arbitrary as an emission species to track the dispersion of the plume. This is because the measurements used for the emission estimate are aerosol optical depth measurements. The chemistry is not actually relevant and in retrospect the detailed description given is confusing. In the revised paper this section has been cut and reorganised to concentrate on the discussion of the relevant modelling of the dispersion of the smoke plumes from the Canberra fires.

3. Page 984: The estimate of the "double-counting" of emissions used in the model run is central to the uncertainty in this analysis. If I read correctly, the emissions used on "day 2" of the simulation are based on the observed enhancement for that day. Some of what is in that enhanced amount is due to CO left over from "day 1" emissions. So the simulated "day 1" distribution of emitted CO are used to correct day 2 a posteriori. The method is to take the remaining "day 1" CO and multiply by the fraction of MODIS grid boxes with AOD values. It's not clear to me what this means or whether it makes sense. I think what is meant is to take the integrated CO of the "day 1" tag in the region and multiply by the fraction of MODIS grid boxes with AOD above the threshold value of 0.2. But this doesn't make sense because I think it implies that that remainder CO is uniform in its distribution (that is, the amount is the same in all the relevant grid cells). What makes sense to me is to screen the "day 1" model remainder CO in the region to retain only model grid boxes (or fractions thereof) that are represented in whatever MODIS AOD grid boxes meet your threshold criteria. The total of all of that CO is then the amount that you would double count as you applied your "day 2" emissions. I think the assumption here is that the model has perfect transport, which we know is wrong but maybe not too wrong.

The referee's interpretation of what we did in the calculation is correct as is the assertion that this makes an assumption that the CO is uniformly distributed. (At least it

assumes that the non-uniformity will not be more or less likely to coincide with where the satellite has good measurements – in which case any bias introduced by the uniformity assumption might be expected to cancel out over the time of the fires). We agree with the referee that the alternative suggestion of screening the model output for previous emissions only in the actual grid boxes with enhanced AOD is probably more sensible. We have re-calculated the double-counting estimate with this method in the revised paper. At the same time we took the opportunity to build an appropriate half-life representative of AOD in the atmosphere into the model output so that the sum of all previous emissions could be made. The double-counting estimate is reduced by this change and so the final emissions estimate is larger. The changes also result in an increased estimate for the negative bias that results from missing data because there is a reduced chance of detecting the smoke at a later date. This too acts to increase the final estimate. For CO the revised emissions estimate is 15 +/- 5 Tg. The relevant sections of the paper have been re-written and updated Figure 3 and Tables 1 & 2 are provided reflecting these new calculations.

4. Page 985, Line 19: I think you mean Eqs. 3 and 4.

Corrected.

5. Page 986, line 14: it should be clarified that when you say larger particles have longer lifetimes you're talking about sub-micron particles as large compared to nano-particles. I know, this is evident in the subject of the paper, but I'm a dust guy and the sentence just reads weird as written.

OK – now re-written as "with lifetimes increasing for higher altitudes and for larger particles (on the sub-micron to nano-particle scale)"

6. Page 989: I'm curious about the background and threshold AOD values chosen. Why didn't you consider the case where background and threshold were both 0.11?

The background value is the average value for days when there is no unusually large impact from fires and this value is 0.11. Because AOD is never entirely uniform there must be some AOD values above this average value on days when there are no nearby fires. Thus choice of the average value to be the threshold value must result in wrongly assigning an emission when there are no real fires present. i.e. this would result in the interference from normal levels of other aerosols like anthropogenic aerosols and sea salt aerosols. The threshold should be chosen to ensure that only significant enhancements of AOD above those normally seen on days without fires are included. This is the mechanism by which other types of aerosols are screened out. Only in the case of an unusual pollution event from one of the other sources (like a dust storm) would there be problems distinguishing the fires from the other greatly enhanced aerosols. The relevant part of the revised paper now reads: "There are sources of enhanced AOD other than fresh smoke from the active fires and so we must be careful not to overestimate the emissions by including AOD that results from dust, sea salt aerosols or aged smoke from other fires. For this reason we impose a threshold value that is set at the maximum AOD measured in the region before the start of the large vegetation fires. By excluding all grid boxes with AOD values no higher than the maximum measured before the fires, we hope to screen out all aerosol sources other than the fires. 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 the 7th January before the fires started was 0.2, and so this was taken as a threshold value to identify fresh smoke. By excluding all values below this threshold we can avoid wrongly assigning a biomass burning emission to an enhancement in AOD that is caused by normal amounts of raised dust, sea salt aerosols or aged smoke."

Response to Anonymous Referee #2

General Comments:

(A) The paper describes a new methodology do estimate trace gas emissions from vegetation fires by using satellite Aerosol Optical Depth (AOD) data from NASA's MODIS

instrument and the chemical transport model MOZART-4. A previously established and published correlation between observed tropospheric AOD columns and correspondent carbon monoxide amounts (CO) was applied in a case study of the Canberra fires in 2003 to demonstrate the performance of this new approach. A whole section of the paper was dedicated to assess the uncertainties of all the method's components and underlying assumptions, which is very useful for readers who are eventually interested in using this method, however it lacks a better foundation of the decisions that led to using the final uncertainty percentages.

See reply to Referee 1- point B.

(B) I am in doubt if the presented method is an alternative to the conventional ways of calculating fire emissions, as it seems to be somewhat complex to reach the final estimate, after performing all the necessary corrections using a chemistry transport model, and uncertainties are not considerably reduced. I wonder if the first, simpler method that included double counting wouldn't be the better approach, given its simplicity and given the fact that the total amount of emissions lies within the uncertainty range of the final result, after all correction work? It is true that the first simpler method may be more useful and in this instance the double-counting and uncertainty analysis yielded a result that encompassed the original estimate without double-counting. As long as the region over which AOD enhancements are counted is not too large given the usual wind fields then the method without correcting for double-counting may well be a better and simpler approach. The error produced by uncorrected double-counting may compensate for smoke missed due to cloud interference. In the revised uncertainty budget these terms do actually almost cancel and this has been pointed out in the revised paper.We believe that the exercise of calculating double-counting was necessary in order to quantify the likely impact of this uncertainty on the method. The fact that in future the method could be applied without such a correction in order to simplify the process is a helpful & positive point from the referee. We have added the follow comment to the paper:

“It is worth noting that the combined effect of correcting for missing data and double-counting does not change the final emissions estimate significantly within the uncertainties of the calculation. This suggest the possibility of neglecting these complicated corrections and using instead a simple addition of the daily enhancements of trace gases inferred from MODIS AOD measurements. Judicial choice of the area over which the calculation is made can ensure that the typical meteorological conditions does not result in circulating wind fields that would lead to excessive double counting. This simplified method assumes that to a first order approximation the effects of double-counting and missing data will approximately cancel.”

(C) I agree with the authors that in fact for distinct fire events the method may be used as an additional tool to derive independent emission estimates, given the fires occur in regions not polluted with a variety of other emission sources (mineral dust, urban/industrial pollution, sea salt, biogenic emissions).

Again we would like to point out that only unusual enhancements of other emission sources of aerosols above the chosen threshold value will cause a problematic interference. The fact that the method is suitable for distinct fire episodes and not for global emissions has been reiterated in the revised paper. (See reply to Referee 1- point A).

Specific Comments / Corrections:

1. p. 978, l. 5, abstract: I found the sentence on the double counting confusing in the abstract. Only later, while reading the details I understood the meaning. Consider removing or rephrasing.

OK – sentence removed.

2. l. 12: “emission factor of carbon monoxide” could be more generally written as “trace gas emission factor”

OK – done.

3. p.980, MODIS AOD data description: You should state that the MODIS AOD product

is AOD at 550 nm and that you used the combined land/ocean collection 5 data.

OK – clarified on page 981 when description of what data we used is first mentioned.

4. p.981 l.17: Why didn’t you use the MODIS AOD “Aerosol Type” variable to identify smoke aerosols?

The use of a threshold value rather than Aerosol Type makes more sense since we also need to filter out aerosols from aged smoke plumes. To clarify this point further the sentence has been re-written: “There are sources of enhanced AOD other than fresh smoke from the active fires and so the background must be chosen carefully so as not to overestimate the emissions by including AOD that results from dust, sea salt aerosols or aged smoke from other fires.”

Later (after explanation of the threshold value for AOD) we have also added: “By excluding all values below this threshold we can avoid wrongly assigning a biomass burning emission to an enhancement in AOD that is caused by normal amounts of raised dust, sea salt aerosols or aged smoke.”

5. l.26+: Suggestion: To express the AODexcess variable mathematically and resume the text above, you could call the determined background values as AODbg = 0.11, and write: “For all AODavg > 0.2 : AODexcess = AODavg – AODbg”. This will make the formula on p. 982, l.10 easier to understand.

Yes – now re-written as: The mean AOD value for a 1° by 1° grid box on the 7th January before the fires started was 0.11 and this was used as a “background” value (AODbg = 0.11). In the subsequent days, whilst the fires burned, the excess AOD that resulted from fresh smoke from the active fires (AODexcess) was estimated using Equation 1

Equation 1 For all AODavg > 0.2 : AODexcess = AODavg – AODbg where AODavg = averaged AOD data in each 1° by 1° grid box

6. p.983, ls 7+8: Comment: At the same time you may have a considerable AOD omission in the MODIS product, since smoke plumes may be mistaken as clouds and

be thus removed from the final product. This would be a counter effect. The sum of both is most probably not zero, though. But the contrasting effect should be mentioned as well. Agreed –this point was alluded to lines 10-13, but we have re-written this to emphasise this point and to build towards a conclusion that the simpler method (without the complex double-counting estimate may actually be more useful).

"Thus the simple addition of daily enhancements in mass of CO may yield an overestimate of the emissions from the fires due to double counting of non-dispersed smoke. At the same time a significant amount of excess AOD may go undetected for a number of reasons including:

1. Smoke plumes may be mistaken as clouds and be thus removed from the final MODIS AOD product. 2. Real clouds (either generated by the fires themselves or present in the region of the fires) cause interference with the retrieval of the MODIS AOD product and result in missing data. 3. The MODIS algorithm may reject data due to other interferences such as sea-glint. 4. Smoke may be transported out of the active region before it is detected by the overpassing satellite

This effect of missing enhanced AOD data will act to underestimate the total emission from the fires and so may counter-act the over-estimate from double counting. However there is no reason why these two opposing effects should be of the same magnitude and so the sum of both is most probably not zero. Moreover, whilst it is not possible to quantify the magnitude of error caused by missing data, a chemical transport model may be used to model the dispersion of the smoke plumes and estimate the amount of double counting. "

7. p.986, l2.: I got a bit lost starting from here. I understood the general idea, of why you are using a chemistry transport model, but I have the impression the details should be presented in a clearer way.

Following the suggestion from referee 1, the model output is now screened for previous emissions only in the actual grid boxes with enhanced AOD. There is now a new

description of the double-counting estimate (that we hope is clearer!) in the revised paper.

8. p.988, l.5: Isn't the MODIS AOD at 550nm? l. 21: Why do you think half the value can be assumed?

Corrected.

9. p.989, l.7: reference?

Following reference added: Remer, L. A., Tanré, D., Kaufman, Y. J., Ichoku, C., Mattoo, S., Levy, R., Chu, D., Holben, B., Dubovik, O., Smirnov, A., Martins, J. V., Li, R.-R., and Ahmad, Z.: Validation of MODIS aerosol retrieval over ocean, Geophysical Research Letters, 29, 8008, 2002.

10. p.990, l. 11: Why 10%?

This value comes from half the double-counting estimate. The choice of this value has been explained in the revised text: "Many of the uncertainties inherent in this method are difficult to quantify because we do not have detailed knowledge of the true variance of the parameters. Despite this fact the exercise of constructing a full uncertainty budget can serve a useful purpose. Where there is no data on which to formally quantify the uncertainties we can make an educated guess that at the probable level of uncertainty in each component will not exceed half its value in the calculation. By this means it is possible to determine how the uncertainty propagates into the final uncertainty in total emissions. This allows us to make a very approximate estimate of the total uncertainties but also it can elucidate which components are likely to dominate the uncertainty budget."

11. p.991, l.22: insert "are" after ": : :, there"

OK – done.

12. l. 30ff.: From what I understood, I don't think the method can be used for estimates

on a large scale. First, the effort for preparing the data for corrections and running a CTM seems to be too complex to make this directly feasible. Second, the presence of other emission sources than fires will jeopardize the quality of results. Should I be wrong about that I suggest clarifying in the text that these issues are not a problem and why

The term “large scale” is liable to misinterpretation. In the revised paper we have used the term “regional scale”. Also we have included a discussion about the potential for using a simplified method without double-counting (as mentioned above). Again the presence of other emission sources will not be a problem unless there is an unusual pollution event from one of these. Clearly the larger the region that is used the more likely an interfering pollution event will occur and the more likely that the meteorology will result in some smoke not being transported out between satellite overpasses. This point has been added to the revised text:.

“Judicial choice of the area over which the calculation is made can ensure that the typical meteorological conditions does not result in circulating wind fields that would lead to excessive double counting. The larger the region that is used the more likely an interfering pollution event will occur and the more likely that the meteorology will result in some smoke not being transported out between satellite overpasses. Thus the area chosen should be as small as possible whilst still containing the vast majority of the smoke emitted during the previous 24 hours.”

13. p.993, l.10; : : :and 347 Tg CO global total for the year 2000 from Hoelzemann (2006), see reference below

OK – re-written as: “and in separate studies for the year 2000 as 496 Tg CO yr-1 (Ito and Penner, 2004) and 347 Tg CO yr-1 (Hoelzemann, 2006).”

14. p. 993, l.15: a newer version of this model with better underlying data for Australia delivers 25 Tg CO/yr for the year 2000 (Hoelzemann (2006). <http://www.mpimet.mpg.de/fileadmin/publikationen/Reports/BzE_28.pdf>

OK – reference changed and subsequent sentences re-written. 15. p.993: please correct “Shultz” three times: The good man’s name is “Schultz”.

Sorry! – corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 977, 2010.