AN EMISSION BUDGET FOR DIOXINS FROM CROP AND BUSH FIRES IN AUSTRALIA.

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Abstract

Previous inventory estimates of dioxins (PCDD), furans (PCDF) and dioxin-like PCB emissions from bushfires in Australia have been limited by the small number of published estimates of emission rates. However, the situation has been improved by a recent field study that measured emission factors for PCDD, PCDF and PCBs from 20 prescribed fires, wildfires and sugar cane fires across Australia. These data are used in a revised inventory methodology that also accounts for uncertainties in inventory activity data and parameters. Incorporating these new data into an emissions budget indicates a new best estimate for Australia's PCDD, PCDF and PCB emissions from open burning in 1994 of 142 g TEQ with an uncertainty range of 31g to 494g TEQ which is 70% lower than the previous estimate. Forest fires and crop residue burning accounted for 8% of total emissions with the remainder occurring in savanna woodlands of northern Australia. Total emission increased by 70% between 1990 and 2001 to 229 g, due entirely to increased fire activity in the savanna woodlands.

Introduction

The source of dioxins (PCDD) furans (PCDF) and coplanar PCBs in Australia continues to be a subject of discussion, particularly the role of forest fires. Emission estimates[1] using the UNEP[2] tool kit have bushfires as a key component, contributing more than 80% of the total Australian dioxin emissions. In contrast, the work of Prange et. al.[3], indicated that PCDD, PCDF and PCB concentration in bushfire smoke plumes is substantially lower than predicted from earlier, laboratory studies of biomass combustion and concluded that forest fires are unlikely to be the major source of PCDD in Queensland. This bimodal view of the strength of the bushfire source poses a major problem for inventories which usually require average, or at least "best estimate" of emission factors for each emission source in order to derive a single total emissions estimate for the nation. The UNEP toolkit uses the average of the published emission factors, noting that there is very a large and unquantified uncertainty in the parameter. The Australian emission inventory prepared in 2002[1] addressed the issue by reporting a lower and an upper estimate for bushfires, of 70 g TEQ and 1700g TEQ respectively, producing, in effect, two national inventory emissions estimates.

The PCDD, PCDF and PCB emission factors have recently been investigated in a field study of open fires comprising prescribed (fuel reduction) fires, wildfires and sugar cane fires in major forest and savanna woodland and cropping regions of Australia [4,5,6]. The dual tracer technique used to determine the emission factors in the field avoids PCDD, PCDF and PCB formation by high temperature surface reactions, which is a potential risk in laboratory tests [4]. This data set is sufficiently coherent to support a new inventory analysis for Australia.

Variability in PCDD, PCDF and PCB formation rates in fires is not the only source of uncertainty in inventory estimates; there is also uncertainty in the other parameters of the inventory algorithms, particularly fuel loads, fire areas and combustion efficiencies. Uncertainty analysis using analytical methods is generally impractical for complex inventory calculations; however, the recent development of Monte Carlo analysis packages for personal computers has changed the situation. In this paper we use the new emission factor data set and a Monte Carlo

analysis that incorporates uncertainty estimates of all other parameters and activity terms to calculate new estimates of PCDD, PCDF and PCB emissions, with uncertainty limits for open burning in Australia.

Materials and Methods

The methodology used here is modified from Australia's National Greenhouse Gas Inventory (NGGI) Methodology [7] used to calculate uncertainties in greenhouse gas emission estimates for prescribed burning of savannas, and prescribed and wildfires in forests.

The emission of PCDD, PCDF and PCB congeners (E_i) from prescribed fires or wildfires is the product of their respective fire areas (A), fuel loads (FL), burning efficiencies (BEF), carbon content (C_c) and emission factors (EF_i) i.e.

$$E_i = A \times FL \times BEF \times C_C \times EF_i \qquad 1$$

BEF is defined as the fraction of total fuel carbon within A that is volatilised, and accounts for both patchiness of the fire (i.e. the fraction of A exposed to flame) and the fraction of fuel exposed to flame that is volatilised.

Emissions from crop residue burning are estimated using a variant of equation 1 in which the fuel burned is estimated as the product of the crop production (P), residue to crop ratio (R:C), dry matter content (DM) and the fraction of crop burned FB, i.e.

$$E_i = P \times R:C \times DM \times FB \times BEF \times C_C \times EF_i \qquad 2$$

All factors other than the PCDD/PCDF and PCB emission factors were sourced from the National Greenhouse Gas Inventory for 2001[8]. The distributions and relative uncertainties are those used in the NGGI to estimate emission uncertainty ranges. The form of the probability distributions for these parameters were selected following IPCC Good Practice Guidance[9]; the distribution means are drawn from the NGGI methodology [8], and the variances are estimated using conservative expert judgement. The emission factors summarised in Table 1 are sourced from the companion paper [5] and are assumed to be lognormally distributed. There were no measurements made for field burning of cereal residues in this study and because the laboratory test yielded qualitatively different results it was decided to apply the emission factors from the field measurements of sugar cane fires [6] to cereal crops.

Table1. Mean PCDD, PCDF and PCB emission factors for open burning in Australia. Units are (g TEQ emitted) (g fuel carbon burned)$^{-1}$. Numbers in brackets are standard deviations

Species	Cane	Prescribed	Wildfire	Savanna
PCDD	1.77 (1.1)	1.23 (0.89)	0.65 (0.76)	2.03 (2.88)
PCDF	0.14 (0.13)	0.36 (0.48)	0.13 (0.15)	0.16 (0.14)
PCB	0.07 (0.04)	0.18 (0.11)	0.13 (0.1)	0.07 (0.04)
Total	**1.98 (1.26)**	**1.77 (1.32)**	**0.91 (1.01)**	**2.26 (3.05)**

The inventory was calculated by a Monte Carlo simulation. For the most part, parameters are assumed to be independent with two exceptions. BEF is assumed to be weakly correlated with fuel load with a correlation coefficient of 0.5 and the emission factors for individual congeners were found to be weakly correlated within fire classes. These correlations are explicitly included in the uncertainty model.

Analyses are run using @RISK Version 4.5 (Palisade Corporation, NY, USA) using Latin hypercube sampling of the PDFs with 3,000 iterations per simulation. Output variables typically converged to less than 1% change of the mean and standard deviation in 500 to 1,000 iterations, which is within the tolerance appropriate for this study.

Accurate determination of the probability density functions for any of the parameters in Equations 1 and 2 is clearly limited by the paucity of published and reviewed data. However, the combination of a Monte Carlo uncertainty analysis and conservative estimates of parameter variance based on extensive field experience ensures that the uncertainties are aggregated correctly through the inventory and that the uncertainty ranges are unlikely to be underestimates.

Results and Discussion

The 2002 dioxin and furan inventory [1] was calculated using activity data for the 1994 inventory year. Using the measured field emission factors from Table 1 in our uncertainty analysis we estimate the total emission of dioxins, furans and dioxin-like PCBs in 1994 was 142 g TEQ with a 95% confidence range of 31 to 494 g TEQ (Table 2). This overlaps the previous inventory range of 72 to 1708 g TEQ however, because the latter is essentially a uniform distribution with no central estimate, comparison between the two inventories can only be made at the extremes of their ranges. The comparison is also somewhat problematic, considering that the confidence levels of the 2002 inventory are undefined. Nevertheless, the impact of the field measured emission factors is to translate the emission estimate distribution to the lower end of the 2002 range. The 2002 inventory was calculated using I-TEQ while the current inventory is based on TEQ-WHO98. Calculating the new inventory using I-TEQ , the emission estimate for 1994 is 126 g I-TEQ (31-428 g).

Almost 84% of the emission occurs in the savanna woodlands of tropical northern Australia and arid zone grasslands of central Australia; wildfires and prescribed fires in the southern temperate forests account for 9% and 4% respectively with the remainder produced from the burning of crop residues in the field. This distribution is similar to the lower bound of the 2002[1] estimate but is a major shift from the upper estimate in which savanna fire emissions comprise only 72% of the total. The difference in the distribution of emissions between fire classes resides in the relatively low emissions factors for the southern Australian forest, particularly wildfires in comparison to the savanna emission factors.

The very low emission factors for wildfires were unexpected because high temperature wildfires were assumed to be a more active PCDD and PCDF production sources than the cooler prescribed fires. Prange et al.[3] and others suggest that PCDD, PCDF and PCB detected in smoke may have been released by volatilization from the soil pool rather than produced *de novo* during combustion. The low concentrations of PCDD and PCDF we observed in the extensive wildfires in NE Victoria in 2003 in comparison to concentrations in smoke from prescribed fires in forests could be consistent with this view. Intense wildfires, particularly crown fires, spread quickly producing less heating of the surface soil than the lower intensity but slow-moving prescribed fires which are confined to the fuel load on the forest floor.

Finally, while limited in number, the new emissions data suggest that that potential exposure of the Australian population by PCDD and PCDF from forest fires may have been significantly overestimated. Most of the Australian population and agricultural production is located on the SE seaboard and the SW region of Western Australia, in regions which are impacted by crop residue fires and forest fires but not savanna fires. The observed emission factors for forest fires in these areas are very much smaller than for coastal Queensland and coastal savanna woodland. It appears likely that the 2002 inventory [1] has overestimated emissions in these areas from 2 to 20-fold.

This study has clarified some of the uncertainty on PCDD and PCDF emissions from fires in Australia, and points to significant regional differences. These questions of regional variability and volatilization from surface soil are the subjects of a study currently in progress.

The trend in national dioxin mass emissions from open burning is presented in Figure 1. In 1990, total annual emissions of PCCD/F and PCBs were approximately 46.5 kg. By 2001, these emissions increased 67% to 76.1 kg caused largely by an increase in fire activity in the savanna woodlands and arid grasslands since 1995 which appears to be associated with a substantial increase in annual rainfall [10]. In terms of mass, emissions in the savanna woodlands are dominated by OCDD, however despite relatively low toxicity of OCDD, total TEQ emissions also increased by 67% between 1990 and 2001 from 140 g TEQ to 233 g TEQ. Emissions from forest fires and crop residues fires decreased slightly over this period therefore the increase in emissions were largely confined to the unpopulated regions of northern and central Australia.

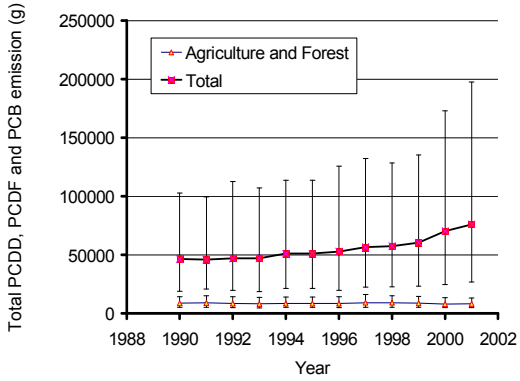

Figure1. Trends in emission of PCDD, PCDF and PCB mass from 1990 to 2001. Error bars indicate the 95% confidence ranges.

Table2. Total emissions of PCDD, PCDF and PCB from open burning in Australia in 1994

	Emission (g TEQ)		
	This study		EA, 2002
Crop residue	3.2	(1.8 – 5.6)	3-260
Prescribed fires	3.6	(1.4-7.9)	
Wildfire	4.9	(1.2 – 15)	7-400
Savanna	130	(20 – 476)	62 – 1240
All	**142**	**(31 – 494)**	**72 – 1700**

In summary, therefore, we conclude that PCDD, PCDF and PCB emissions in Australia were substantially overestimated by the 2002 inventory. Our current best estimate is 8% of the 2002 inventory upper limit; however, the uncertainties remain large. Emissions have increased substantially since 1990 but the increase is confined to the largely unpopulated regions of tropical northern Australia and appears to be associated with climate variation rather than anthropogenic activity.

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