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An Investigation of the Variability of Particulate Emissions from Woodstoves in New Zealand

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ABSTRACT

Woodburning is the dominant source of PM_{10} in most of New Zealand, and the principal cause of exceedances of the national air quality 24-hr standard (50 µg m^{-3}). Over the last decade New Zealand researchers have used multiple techniques to characterise woodstove emissions, including seven campaigns to measure real-life PM_{10} emissions from in-situ woodstoves. All measurements have shown considerable variation, both within and between campaigns. PM_{10} emission factors from in-situ tests exhibit a log-normal distribution with a geometric mean (± standard deviation) of 9.8 g kg^{-1} (± 2.4 g kg^{-1}) and 3.9 g kg^{-1} (± 3.8 g kg^{-1}) (dry wood) for older and low-emission stoves respectively. Since the distribution is log-normal we recommend the use of geometric mean rather than arithmetic mean as the emissions factor used for inventories. This paper examines the variability of PM_{10} emissions from woodstoves using a Kernel Density Estimate to compare the distributions within and between campaigns and correlations between coefficients of variations (c.v.) to see whether the variation in emissions factors could be reduced by the collection of further measurements to increase the size of the total dataset. We conclude that variation in emissions is inherent in the way woodstoves are used in real-life and that regulators will need to allow for a range of emissions in management plans as a definitive emission factor may not be possible.

Keywords: Woodsmoke; PM_{10}; Emission factors.

INTRODUCTION

Use of solid fuel, mostly wood but also coal, is a major source of night-time airborne pollution in many places in New Zealand, showing clear diurnal patterns (Anclett *et al.*, 2012; Mitchell, 2012). The problems of woodburning in NZ are similar to those in alpine valleys across Europe and North America where woodsmoke collects under night-time inversions, leading to high concentrations of particles.

The 2013 census found that 546,000 dwellings (36 percent of all dwellings) in New Zealand used wood for heating (MfE, 2014) a similar number to small northern European countries such as Norway or Denmark. A 2005 survey (MfE, 2005) estimated that these fires could burn more than 13,000 tonnes of wood a day during winter.

The country has a National Environmental Standard (NES) for PM_{10} of 50 µg m^{-3} in air as a 24 hour average. Most breaches of this standard are attributed to domestic heating with 24-hour levels of more than 200 µg m^{-3} having been recorded in some towns in New Zealand. The standard also has design criteria for wood burners. New installations must meet an emission limit of 1.5 g kg^{-1} of particles (approximately equal to 100 mg MJ^{-1} for pine (dry wood)) when tested to New Zealand Standard AS/NZS4013 (AS/NZS, 1999).

Source apportionment studies (Scott, 2005; Davy *et al.*, 2007; Wilton *et al.*, 2007; Davy *et al.*, 2011) have indicated that particles from wood-burning is a major contributor to elevated winter-time concentrations of PM in various locations around New Zealand. Woodsmoke has been attributed to as much as 90% of winter-time PM, both PM_{10} and $PM_{2.5}$ (e.g., Scott, 2005; Davy *et al.*, 2011).

The combustion conditions leading to increased particle emissions from wood burning are relatively well understood (see e.g., Houck *et al.*, 2008 and references therein; Kocbach-Bølling *et al.*, 2009) but emissions measurements are highly variable (Houck *et al.*, 2008; Nussbaumer *et al.*, 2008). The ability of regulatory test methods to represent inter-burner performance on a real-life basis has been raised both in New Zealand and elsewhere (Scott, 2005 and references therein). Houck *et al.* (2008) also point out that many certification standards are over two decades old and in practice should only be considered as benchmarks designed to provide a consistent basis for comparison of appliances recognising that real-life testing is difficult and variable and therefore only loosely predictive of in-home emissions.

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Many studies have been carried out to measure and characterise wood-smoke but they have largely been carried out under laboratory conditions, often with attempts to simulate real-life (Wilton, 2014). Laboratory set-ups are generally large and cannot be transferred to the field, and many of the instruments used do not work well in poor environmental conditions. Consequently, real-life measurements of wood-smoke are rare (Tissari *et al.*, 2007).

A number of studies have been carried out in New Zealand to measure real-life emissions from domestic wood burners. Measurements have been made *in-situ* in volunteers' homes, using the woodstove as they would normally. Measurements were made using a set-up based on Oregon Method 41, also known as the Condar Method (Barnett, 1983) or a method developed by CSIRO (Meyer *et al.*, 2008), both of which involved inserting a sampling tube through the wall of the appliance chimney and sampling direct from the chimney into a dilution chamber. Reportedly the state of repair of the appliances was variable as was the state of the houses themselves (Martin Unwin, NIWA pers comm). The volunteer households were generally chosen as randomly as possible but the size of the pool from which they were chosen and hence how representative they are of the general population is not known. The same parameters were not measured across all campaigns, particularly flue temperature and oxygen flow. Table 1 shows the average emissions factors published from all known in situ woodstove tests carried out in New Zealand. The studies report average emissions in the range 1.4 g kg^{-1} to 11 g kg^{-1} (≈ 100 mg MJ^{-1} to 800 mg MJ^{-1}). In all, 51 woodstoves have been tested in seven different campaigns, 37 "NES compliant" (i.e., type tested to AS/NZS4013), two woodstoves with laboratory emissions ratings between 1.5 and 3.5 g kg^{-1} and 12 pre-1994 woodstoves with no or unknown laboratory emissions ratings. Wilton (2014) gives a summary of emissions testing in New Zealand including comparison of methods and results from the seven studies. Overall, the low emission woodstoves tested do have lower emissions than the smaller sample of older woodstoves. The maxima are as large as the older woodstoves but the minima and mode are lower.

There is considerable difficulty in comparing results from *in-situ* emissions testing in New Zealand to studies carried out in other countries because of different measurement methods, stove types, reporting units and approaches to determining what represents real life operation. Although there are many methods of expressing the results of emissions testing, in New Zealand the units used have generally been grams of PM_{10} emitted per kg of wood burnt (g kg^{-1}) since this is the unit required for emissions inventories and specified in the New Zealand Standard AS/NZS4013. A review by Nussbaumer *et al.* (2008) collected together "official" and other reported emissions factors from across northern Europe. They found that reported emissions factors can vary by orders of magnitude depending on the type of sampling undertaken. The largest effect on reported emissions factors came from whether a dilution tunnel had been used or not. This can increase the quantity of particles measured by more than one-hundred fold with reported emissions factors for dilution tunnel measurements between 200 mg MJ^{-1} and

Table 1. Summary of real-life woodstove emission testing in New Zealand.

Location	Campaign name	Appliance type	Number of appliances	Number of runs	PM emissions (wet weight) (g kg^{-1})	Reference
Christchurch/Nelson	Ch_2005	NES compliant woodstoves (< 1.5 g kg^{-1})	4	28	10.8	(Scott, 2005)
Christchurch/Nelson	Ch_2005	Woodstoves > 1.5 < 3.5 g kg^{-1}	2	15	8.4	(Scott, 2005)
Tokoroa	Tokoroa_2005	Pre-1994 wood woodstoves	12	96	11	(Wilton and Smith, 2006)
Tokoroa	Tokoroa_2006	NES compliant woodstoves (< 1.5 g kg^{-1})	9	59	3.6	(Kelly *et al.*, 2007a; Smith *et al.*, 2009)
Nelson	Nelson_2007	NES compliant woodstoves (< 1.5 g kg^{-1})	18	92	3.3	(Smith *et al.*, 2009)
Taumarunui	Rotarua_2007					
Rotorua	Tamaranui_2007					
Christchurch	Ch_2009	1.5 and 1.0 g kg^{-1} woodstoves	6	100	7.3	(Bluett and Meyer, 2011)

2000 mg MJ^{-1} for conventional woodstoves, with a typical value of 340 to 544 mg MJ^{-1}. The EMEP/EEA air pollutant emission inventory guidebook (EEA, 2013) collates particulate matter emission factors reported in the literature for wood burning from over 50 studies measuring emissions from a variety of installations burning wood for residential purposes. It gives values from 10 to over 1400 mg MJ^{-1} for enclosed stoves but it should be noted that this includes a range of technologies including pellet burners, which are not included in the New Zealand studies. In New Zealand measurements have been made using dilution tunnel methods or equivalents and results are consistent with but on the low side of other reported values.

The original objective for the first measurement campaigns was simply to measure emissions factors for emissions inventories (Scott, 2005; Smith *et al.*, 2009). When considerable variation was found in the measurements, further campaigns were conducted to establish the extent of variation between different kinds of woodstoves and between households. It was expected that the collection of more data would reduce the uncertainty in the mean emissions factors.

This paper investigates how the variability in results can be addressed by aggregating the results from the campaigns and whether, specifically, that variation can be reduced by further, similar measurements.

METHODS

To answer these questions the combined dataset from all the in-situ measurement campaigns in New Zealand was examined. In order to examine the combined dataset from all the different measurement campaigns it is necessary to determine the similarity between the estimated emission factors from different studies and the similarity between the estimated emission factors from different woodburners within each study. If the studies' results are too different from each other, comparisons are not valid as different, often unmeasured, factors may be driving the results. A kernel density estimates (KDE) method was used to compare the pairs of estimated emission distributions. Kernel density estimates (KDEs) provide a data-driven method for approximating length-frequency data with probability density functions (Sheather and Jones, 1991). Like a Kolmogorov-Smirnov (KS) test, KDEs also provide a non-parametric approach to compare pairs of distributions via a permutation test for shape and location of mean or median (Zar, 1999). The KDE approach compares the area between two probability density functions, rather than the point difference used by the KS test, so the KDE test is more sensitive to differences between the pair of distributions (Langlois *et al.*, 2012). Langlois *et al.* (2012) developed R functions implementing the kernel density estimates for length frequency comparison. Those functions were used for our woodburner emission analysis. To investigate differences due to shape alone, emission factor data were standardised by median and variance (y = x-median/stdev), as suggested by Bowman and Azzalini (1997, 2010).

The correlations between coefficient of variations (c.v.) and sample size (number of daily burning runs), number of woodburners, and average sample size of each woodburners were explored to see whether the variation in emissions factors could be reduced by the collection of further measurements to increase the size of the total dataset.

RESULTS AND DISCUSSION

A total of 390 daily burns have been tested from 51 woodstoves in seven campaigns, and the measured emission factors for each day range from 0.2 g kg^{-1} to 90.6 g kg^{-1}. Fig. 1 shows results for all campaigns (top panel) and for the NES compliant burners only, separated into individual campaigns (bottom panel). Some woodstoves have consistently higher emissions than others and some campaigns also report higher measurements (and hence higher averages) but a robust understanding of the variability within individual woodstoves cannot be derived from the emission measurements due to the short duration of the campaigns (all less than three weeks). An individual woodstove (for example the first woodstove in Christchurch 2009 or the second in Tokoroa 2005) can have a range of emissions that are nearly as large as the range of the entire dataset and emissions from the same woodstove can vary by a factor of ten from one day to the next. The emission factors of all seven campaigns have distributions that are apparently skewed to the right, but they have different means and some are wide spread while others are relatively tight (Fig. 2).

If all the seven campaigns are collated, the emission factors measured from all 390 burning tests of the 51 woodstoves have a log-normal distribution (Fig. 3, top). The geometric mean is 4.9 g kg^{-1} with a geometric standard deviation of 3.8 g kg^{-1}. The log-normal distribution holds true for the emission factors when NES compliant and pre-1994 woodstoves are considered separately (Fig. 3, bottom). The geometric mean and standard deviation are 9.8 g kg^{-1} and 2.4 g kg^{-1} for pre-1994 woodstoves and 3.9 g kg^{-1} and 3.8 g kg^{-1} for NES compliant woodstoves. The average emissions factors from the NES compliant woodstoves, the so-called low emissions woodstoves, are indeed lower than the older woodstoves, but the variation is greater amongst them.

Tissari *et al.* (2007) report a range of 0.6 g kg^{-1} to 2.7 g kg^{-1} for PM_1 measured from seven different types of wood stove in real-life. In-situ measurements in Australia (Meyer *et al.* 2008) reported an emissions factor of 10.6 g kg^{-1}, McCrillis (2000) found that PM_1 constituted between 60% and 90% of the total particulate catch in EPA 5H and 5G tests with PM_{10} averaging 94% of the total. Therefore results from Tissari *et al.* (2007) are comparable to the range of New Zealand results. Tissari *et al.* (2007) found that in field studies the emission levels were about up to 3 times those in laboratory studies, which is also consistent with New Zealand findings, Scott (2005) found real-life values up to five times those of simulated real-life and 16 times the type approval tests.

To quantitatively determine the similarity or difference between the distributions of the emission factors, a kernel density estimate (KDE) method (Sheather and Jones, 1991; Langlois *et al.*, 2012) was applied to compare the distributions for both mean and shape. Fig. 4 shows the KDE results of

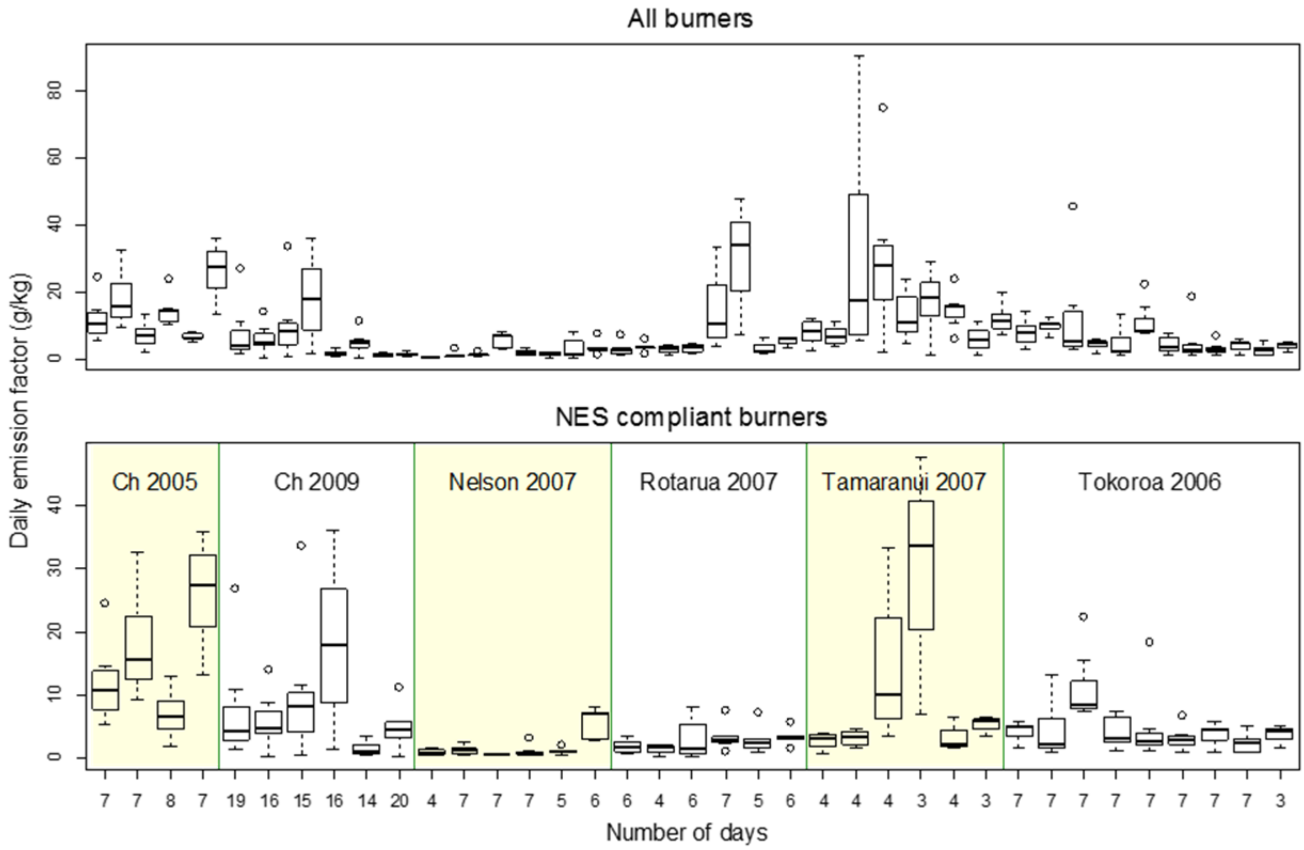

Fig. 1. Results for all campaigns (top panel) and for the NES compliant burners only, separated into individual campaigns (bottom panel).

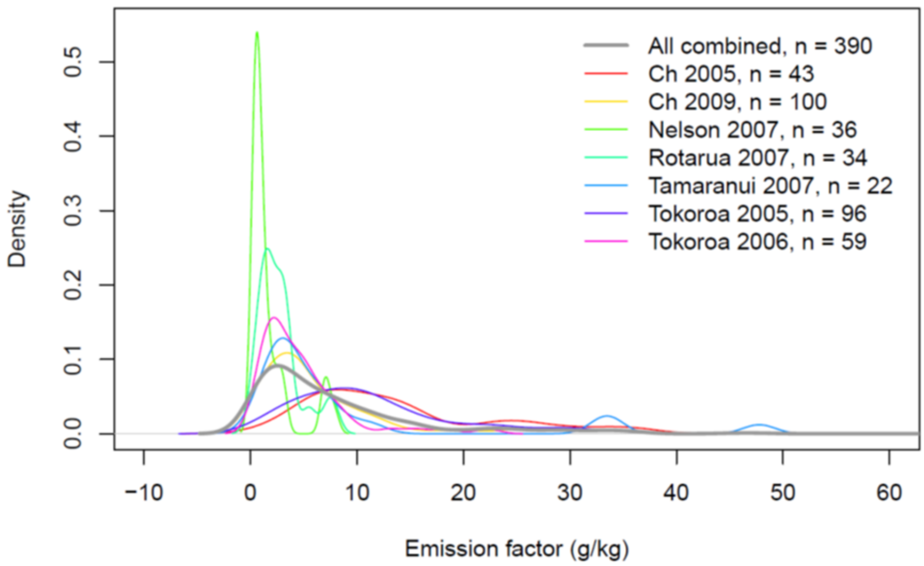

Fig. 2. The woodstove emission distributions of the sample data from the seven campaigns listed in the legend and all data combined.

comparing the means of each campaign with the overall mean and Fig. 5 shows a comparrison of the shape of the distribution of each campaign with the shape of the overall distribution. In order to compare the shape of the distributions, the campaign data are normalised so that the means coincide. While the means vary between campaigns, the shape of the distributions is always not significantly different. Three out of seven campaigns have similar means to the mean of the whole dataset, while others are clearly different (Fig. 4). However, each of the seven distributions has a similar shape

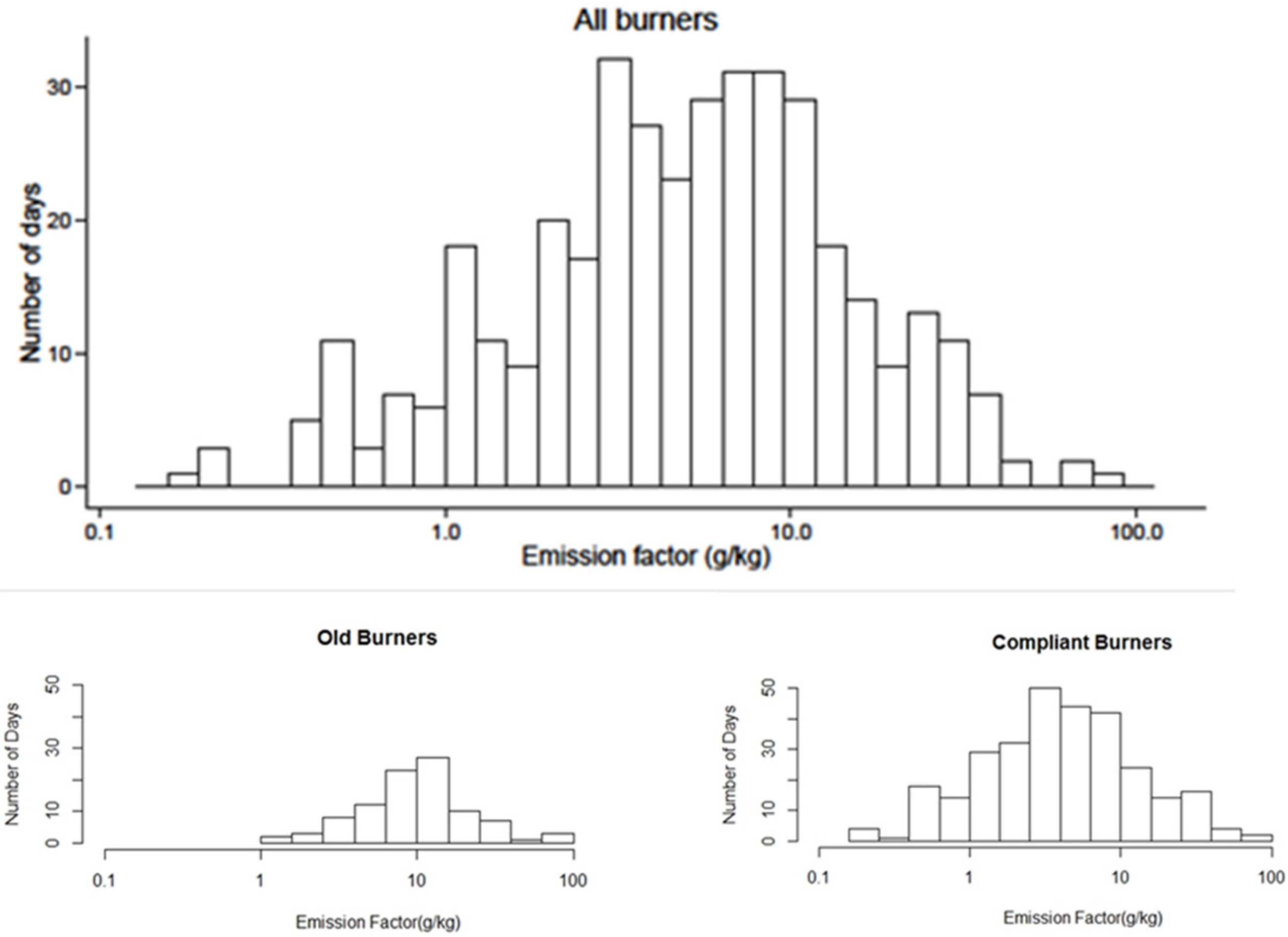

Fig. 3. Log-normal distribution of emission factors measured from all 390 burning tests of the 51 woodstoves (top). NES compliant and pre-1994 woodstoves (bottom).

to the whole dataset's distribution shape, with the lowest p-value of 0.057 (Fig. 5). Therefore we conclude that the woodstoves emissions factors measured in the seven real-life campaigns are log-normally distributed.

Two campaigns were chosen to examine how emission factors measured from individual woodstoves are distributed: Christchurch 2009, which has the highest sample size, and Tokoroa 2005, which includes highest number of woodstoves and has the widest distribution. The individual woodstoves have very variable emission distributions, some of which are very tight, while others are relatively wide (Fig. 6). Many of the individual woodstove emission distributions do not show the skew pattern that can be seen in the campaign distributions whilst others do. This indicates that there is variation not only between but within households that is contributing to the total variation.

It needs to be borne in mind when comparing individual emission distributions that each individual woodstove was tested for only a few days (average 11, max 20, min 7 days for the two campaigns considered here and average 8, max 20, min 3 days across all seven campaigns) and as mentioned above, an individual woodstove can have a range of emissions that are nearly as large as the range of the entire dataset.

Analysis of early campaigns by Scott (2005) and Wilton *et al.* (2006) concluded that whilst some of the variation could be attributed to wood moisture, flue temperature and air flow the most important variable in real-life woodstove emissions is the operator indicating that variables not being measured in those campaigns may be responsible for the variation. Wilton and Bluett (2012a, b) were able to attribute up to half the variation to wood moisture with a further 20% explained with the inclusion of flue temperature and oxygen availability. These results are consistent with laboratory measurements made in New Zealand (Xie *et al.*, 2010; 2012) investigating the effect of several variables (wood species, wood moisture, wood size, make of woodstove and stage of burn – start-up, high burn, low burn) on emissions. A total of 155 runs were carried out using three different woodstoves. If all the runs are aggregated, they show similar distributions to the results from the real-life studies (Figs. 7 and 8). So it is reasonable to assume that the covariate association with the emission for the two dataset should be similar too. Hence the cause of the variance in real-life can be associated with the same variables. An ANOVA with log-transformed response and interaction of factors carried out on these results indicates that wood moisture and burning stage are the largest influence on emission from a stove in this test, which are both parameters under control of an operator.

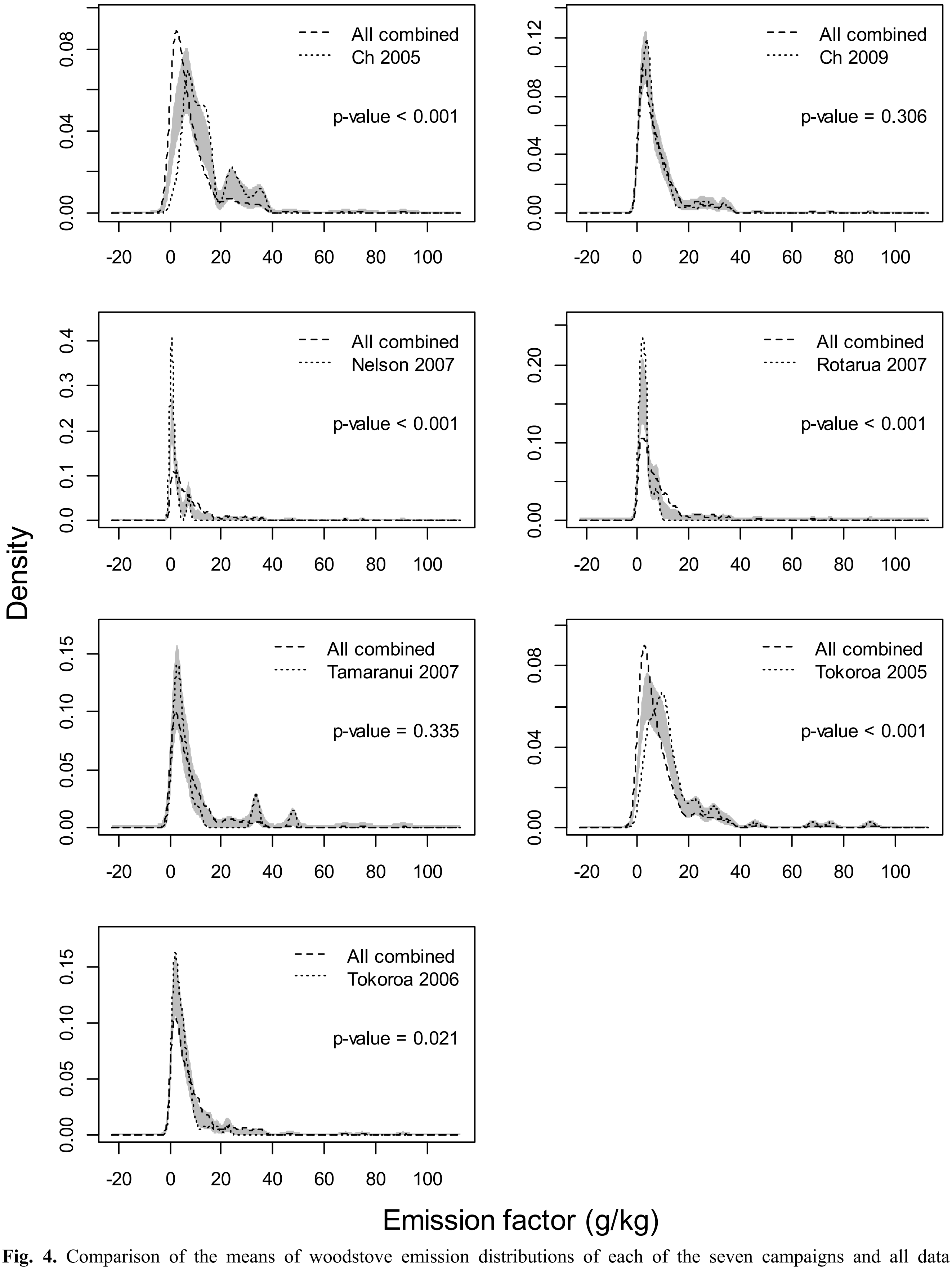

Fig. 4. Comparison of the means of woodstove emission distributions of each of the seven campaigns and all data combined.

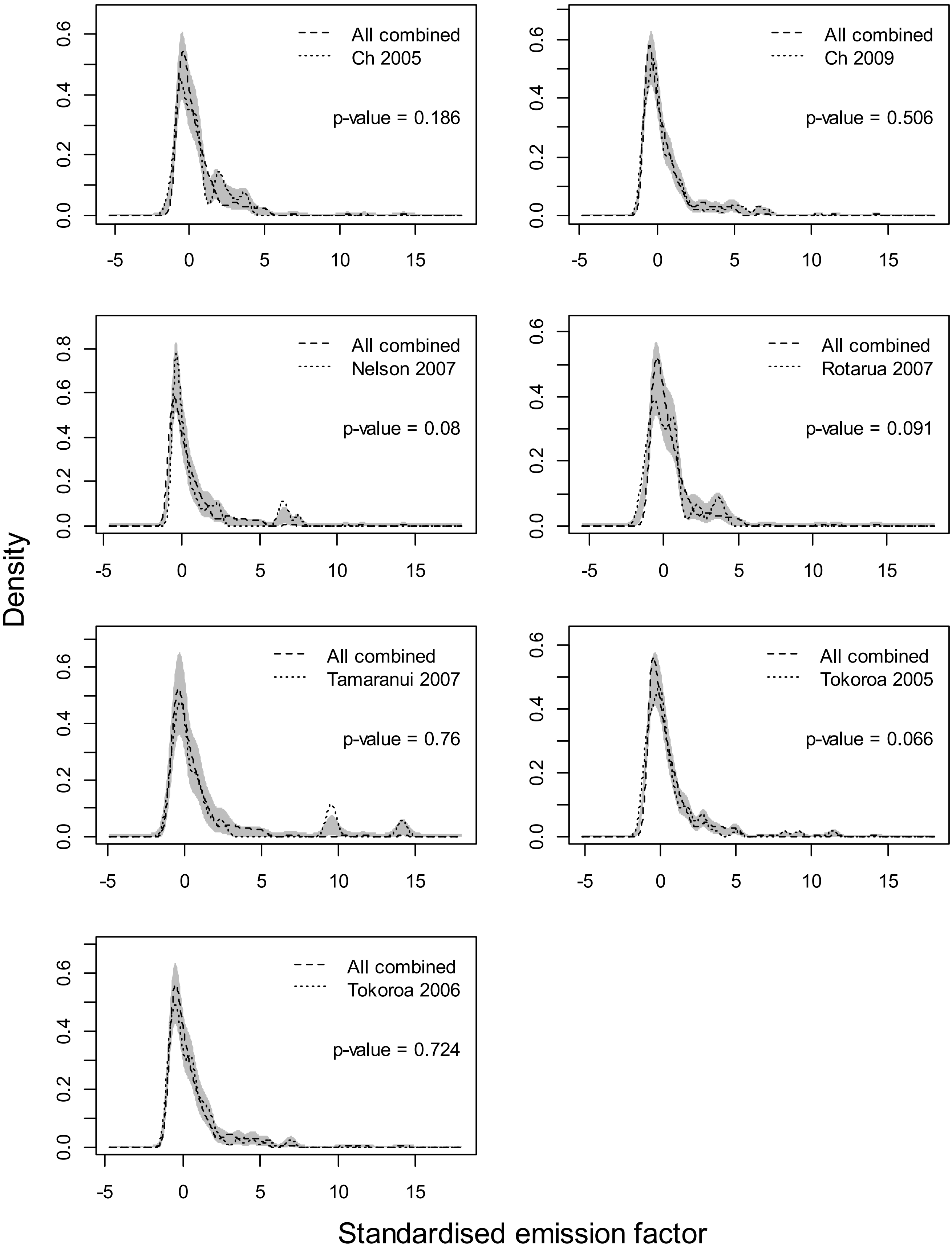

Fig. 5. Comparison of the shapes of woodstove emission distributions of each of the seven campaigns and all data combined.

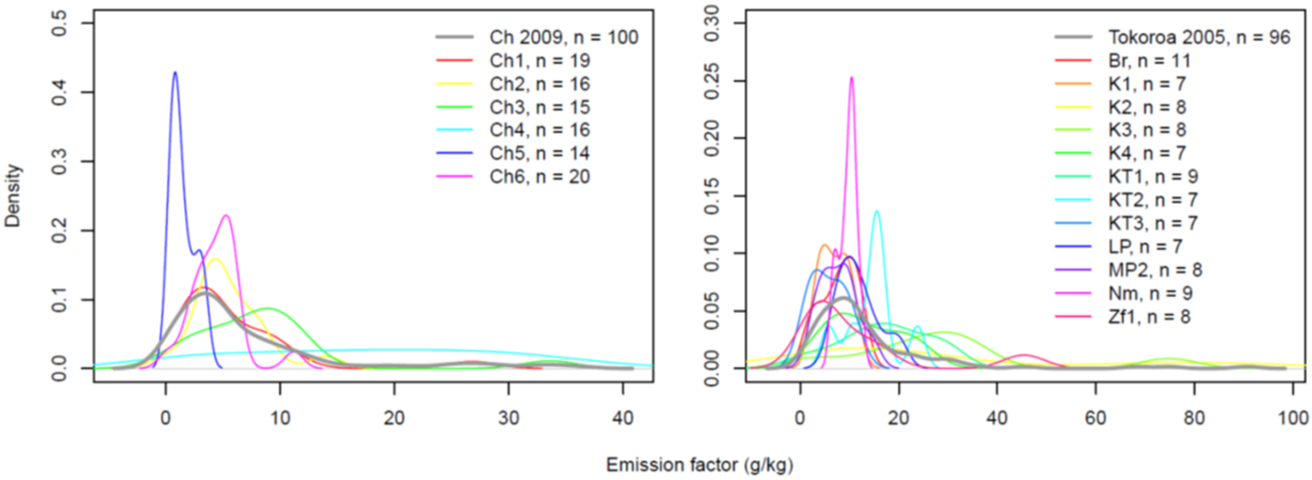

Fig. 6. Woodstove emission distributions in campaigns Ch 2009 and Tokoroa 2005.

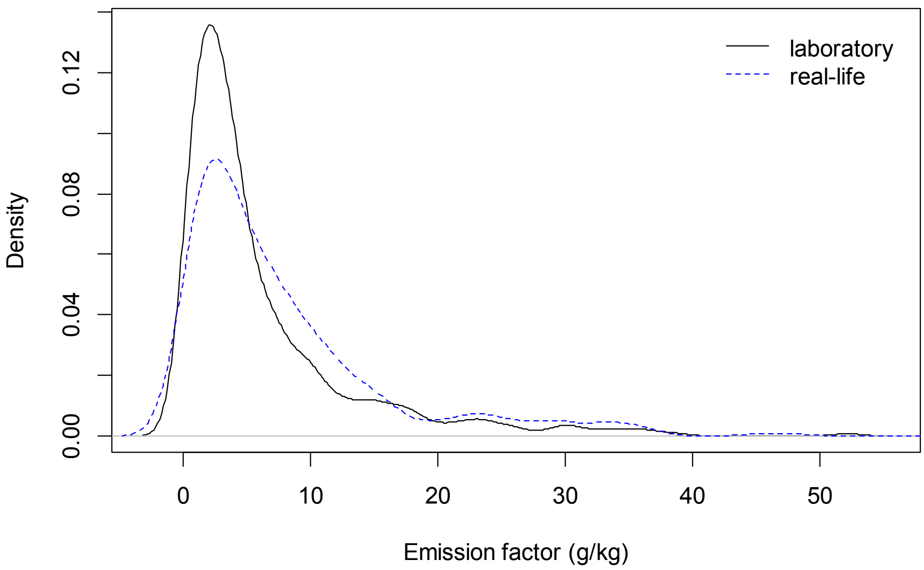

Fig. 7. Woodstove emission factor distributions from laboratory tests in contrast to the seven campaigns of real-life tests.

Scott (2005), Wilton e*t al.* (2006) and Wilton and Bluett (2012a, b) all concluded that further measurement was required to reduce variability by increasing the sample size. However, increasing sample size in a future study may not reduce the variance. We explored emission measurements from the seven campaigns and have not found convincing proof of a relationship between the variances and sample sizes. Some basic statistics are given in Table 2 on the seven campaigns and it shows that all coefficients of variance (c.v.s) are around 1 and that larger sample sizes do not improve the c.v.s. There is almost no relationship between the numbers of tests and c.v.s in the seven campaigns and the same is true of the 51 woodstoves (Fig. 9).

CONCLUSIONS

Over the past decade, seven campaigns have been carried out in New Zealand trying to obtain real-life emission factors for domestic woodstoves. The results have consistently shown considerable variation. It would appear that, in real-life at least, emissions from woodstoves are inherently highly variable as no direct relationship has been found between the variance and sample sizes of the campaigns, which means more measurements in future will not reduce the variation. Regulators will need to allow for a range of emissions in air quality management plans as a definitive emission factor may not be possible.

The covariates associated with the wood burning process, e.g., the wood used, the operation of the woodstove, or environmental conditions, have significant effects on the variation of emission measurements.

Emission measurements from all seven campaigns exhibit a log-normal distribution and when all seven campaigns are aggregated together, the emissions display the same distribution. Aggregated emission factors from laboratory tests of combinations of covariates also yield a log-normal distribution.

Given the apparent long tail of the woodstove emission

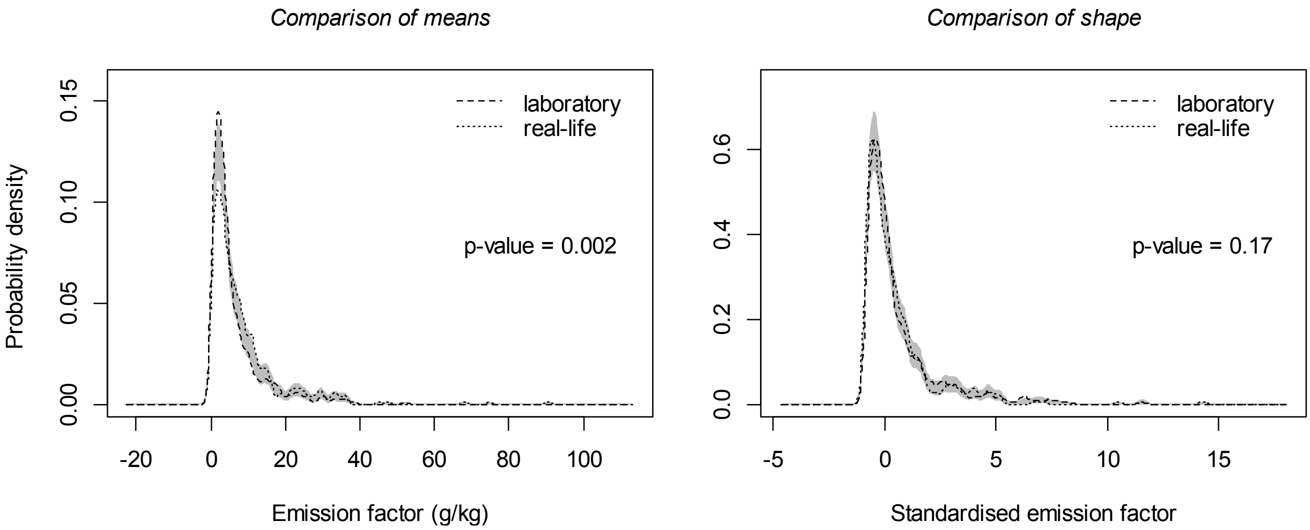

Fig. 8. Woodstove emission distributions from the laboratory tests in contrast to the seven campaigns of real-life tests.

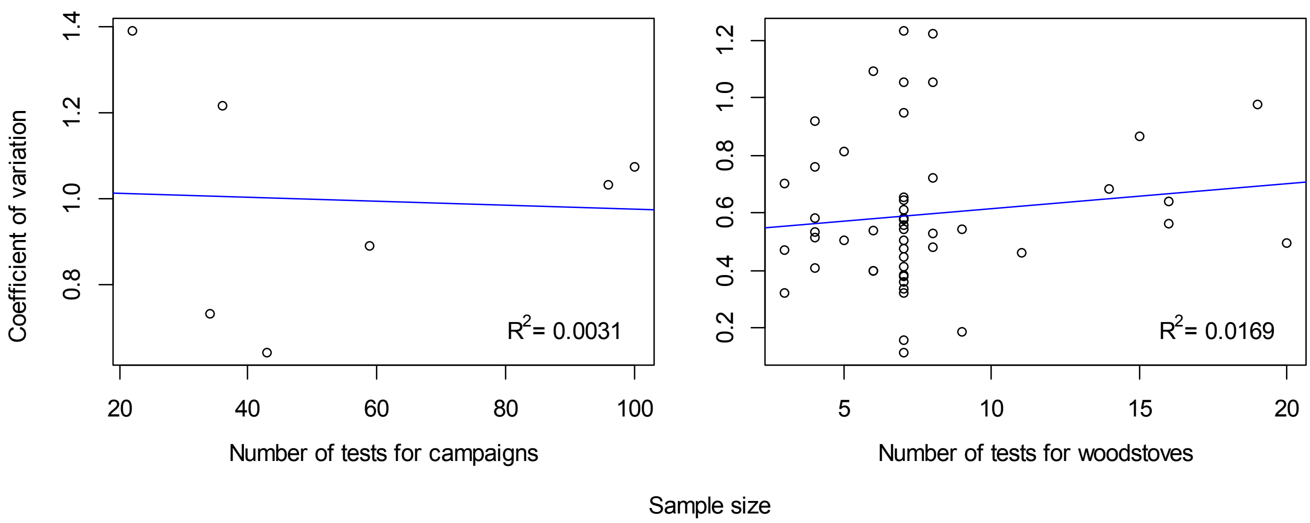

Fig. 9. The relationship between sample sizes and c.v.s for the seven campaigns (left panel) and the 51 woodstoves (right panel).

Table 2. Basic statistics of the eight studies emission results.

campaign	sample size	mean	variance	c.v.
Ch_2005	43	20.82	346.97	0.89
Ch_2009	100	7.27	61.14	1.08
Nelson_2007	36	1.75	4.52	1.22
Rotarua_2007	34	2.71	3.94	0.73
Tamaranui_2007	22	8.94	154.93	1.39
Tokoroa_2005	96	13.82	204.24	1.03
Tokoroa_2006	59	4.74	17.75	0.89

distribution, mean and standard deviation will not give a precise description of the woodstove emissions. The mean will not be the most likely value if a random sample is taken and the mean ± standard deviation will not properly cover the actual asymmetric emission range. The median and 95% confidence interval will provide a more appropriate description of the emission range of the woodstoves on different days in a study. Although the median may be the best value to describe the distribution of the emissions, for emissions inventory purposes the mean is the most useful number as the total emissions in a given area will be the number of woodstoves times the average emissions. In this instance, since the distribution is log-normal we recommend the use of geometric mean rather than arithmetic mean as the aggregate emissions factor used for inventories. Therefore, in New Zealand we recommend using values of 9.8 g kg^{-1} (± 2.4 g kg^{-1}) for older stoves and 3.9 g kg^{-1} (± 3.8 g kg^{-1}) (dry wood) for low-emission stoves.

Further work might include longer time series from individual woodstoves including intervention studies to investigate changes in operator behaviour, which may help understand the day to day variability. Issuing all householders with a standardised batch of firewood may help in comparing one household with another in order to investigate the variability of the appliances.

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