

## Emissions of Carbonyl Compounds from Various Cookstoves in China

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This paper presents a new database of carbonyl emission factors for commonly used cookstoves in China. The emission factors, reported both on a fuel-mass basis (mg/kg) and on a defined cooking-task basis (mg/task), were determined using a carbon balance approach for 22 types of fuel/stove combinations. These include various stoves (e.g., traditional, improved, brick, and metal, with and without flue) using different species of crop residues and wood, kerosene, and several types of coals and gases. The results show that all the tested cookstoves produced formaldehyde and acetaldehyde and that the vast majority of the biomass stoves produced additional carbonyl compounds such as acetone, acrolein, propionaldehyde, crotonaldehyde, 2-butanone, isobutyraldehyde, butyraldehyde, isovaleraldehyde, valeraldehyde, hexaldehyde, benzaldehyde, *o*-tolualdehyde, *m,p*-tolualdehyde, and 2,4-dimethylbenzaldehyde. Carbonyls other than formaldehyde and acetaldehyde, however, were rarely generated by burning coal, coal gas, and natural gas. Kerosene and LPG stoves generated more carbonyl compounds than coal, coal gas, and natural gas stoves, but less than biomass stoves. Indoor levels of carbonyl compounds for typical village houses during cooking hours, estimated using a mass balance model and the measured emission factors, can be high enough to cause acute health effects documented for formaldehyde exposure, depending upon house parameters and individuals' susceptibility.

### Introduction

Carbonyl compounds (aldehydes and ketones) are of concern due to their potential adverse health effects and their environmental prevalence. Several aldehydes are targeted as potentially toxic by the Clean Air Act Amendments of 1990 (1). Reported health effects from exposures to aldehydes include mucosal membrane and eye irritation in animals and humans (2), genetic damage, and cancer (3–5). Carbonyls are present in the atmosphere and play a significant role in

photochemical smog processes in which two criteria air pollutants, ozone and fine particulate matter, can be generated (6, 7).

Atmospheric carbonyl compounds are produced directly from incomplete combustion of biomass and fossil fuels and through the atmospheric photooxidation of hydrocarbons (8, 9). Natural sources also contribute to atmospheric concentrations of carbonyls through biogenic emissions of formaldehyde, acetaldehyde, and a few other aldehydes as well as photochemical oxidation of naturally emitted hydrocarbon precursors, e.g., isoprene (10–12). Various carbonyl compounds have been measured in polluted urban air during photochemical episodes (13–16) and in the “clean” rural air (11, 17).

Some carbonyl compounds are released into occupational and residential indoor air settings from building materials, furniture, and consumer products (18–22) and via reactions between indoor ozone and alkenes (23–26). Cigarette smoke is another significant indoor source of several aldehydes (27, 28).

The majority of outdoor measurements of carbonyl compounds have focused on formaldehyde and acetaldehyde, the two most abundant aldehydes in the atmosphere, while indoor measurements have almost exclusively targeted at formaldehyde which is often present at higher concentrations indoors than outdoors due to indoor sources. Few studies, however, have found other carbonyl compounds present in both outdoor and indoor air. For example, acrolein, crotonaldehyde, glyoxal, and methylglyoxal have been measured in the ambient air of Los Angeles (29); propionaldehyde, 2-furaldehyde, butyraldehyde, benzaldehyde, isovaleraldehyde, *n*-valeraldehyde, and *n*-hexaldehyde have been measured in both indoor and outdoor air of six New Jersey homes (23). Many of these measured aldehydes are toxic or potential toxic (30–33).

Despite the fact that carbonyl compounds are ubiquitous in the ambient atmosphere and indoor environments, few data are available to understand the nature, extent, and sources of human exposures to these compounds. Even fewer data are available on emissions of carbonyl compounds from a very common indoor source throughout the world: cookstoves.

In developing countries, the primary source of indoor air pollution is often the combustion of dirty fuels used for cooking and/or heating, which has been found to be responsible for many indoor pollutants including carbon monoxide, particulate matter, sulfur dioxide, nitrogen dioxide, and various organic compounds (34–38). Although the elevated formaldehyde concentration has been measured in the kitchen air during cooking hours (39, 40), no measurements have been made on other carbonyls associated with household combustion sources.

In a study of greenhouse gases from widely used small-scale combustion devices in developing countries, we have measured emission factors of carbon dioxide (CO<sub>2</sub>), carbon monoxide (CO), methane (CH<sub>4</sub>), total non-methane organic compounds, total suspended particulate matter, oxides of nitrogen (NO<sub>x</sub>), nitrous oxide (N<sub>2</sub>O), etc. for 56 types of fuel/stove combinations in China and India (41, 42). Among these 56 fuel/stove combinations, 22 were also measured for emissions of carbonyl compounds in China. Using a sensitive measurement method, we were able to measure up to 16 carbonyls in the flue gases of the tested cookstoves. We have also measured simultaneously the fuel energy content and

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TABLE 1. Description of the Fuel/Stove Combinations Tested

serial no.	symbol (fuel/stove/flue <sup>a</sup> )	description	
		fuel	stove
1	WR/brick/1	wheat residue	brick stove with flue
2	WR/imp/1	wheat residue	improved brick stove with flue
3	MR/brick/1	maize residue	same as #1
4	MR/imp/1	maize residue	same as #2
5	FW/brick/1	fuel wood	same as #1
6	FW/imp/1	fuel wood	same as #2
7	FW/Indian/0	fuel wood	metal stove without flue (from India)
8	BW/brick/1	brush wood	same as #1
9	BW/Indian/0	brush wood	same as #7
10	HC/metal/1	honeycomb coal briquette	metal stove with flue
11	HC/metal/0	honeycomb coal briquette	metal stove without flue
12	HC/imp/0	honeycomb coal briquette	improved metal stove without flue
13	CB/metal/1	coal briquette	metal stove with flue
14	CB/metal/0	coal briquette	metal stove without flue
15	UC/brick/1	unprocessed coal	brick stove with flue
16	KE/wick/0	kerosene	kerosene wick stove without flue
17	KE/pres/0	kerosene	kerosene pressure stove without flue
18	LPG/trad/0	liquefied petroleum gas	LPG traditional stove without flue
19	LPG/inf/0	liquefied petroleum gas	LPG stove with an infrared head without flue
20	CG/trad/0	coal gas	traditional gas stove without flue
21	NG/trad/0	natural gas	same as #20
22	NG/inf/0	natural gas	gas stove with infrared head without flue

<sup>a</sup> Flue code: 1 = with a flue, 0 = without flue.

the stove efficiency along with the stove emissions. This enables us, in this paper, to report the emission factors both on a fuel-mass basis and on a cooking-task (delivered energy) basis, the latter being particularly useful for comparing the air pollution potential of different fuel/stove combinations and assessing the impacts of fuel/stove substitutions. To illustrate the potential exposures resulting from the cookstoves, we have also related the measured emission factors to indoor air concentrations during cooking hours using a single compartment mass balance model.

## Methods

The 22 fuel/stove combinations tested for carbonyl emissions are described in Table 1. Those tested represent a wide range of household fuels and stoves most commonly used in rural and urban areas of China. Except for those stoves using piped gas fuels, i.e., fuel/stove combinations 20, 21, and 22, which were measured in actual homes, all other fuel/stove combinations were tested in a simulated village kitchen house at Tsinghua University's rural campus about 50 km from Beijing.

**Sampling Method.** Since it is known that emissions from most solid fuels can vary during the combustion process (43), an integrated sampling strategy was employed to collect emissions during an entire burn cycle (from fire start to fire extinction). This provides emission data that can represent a realistic burning situation. In addition, it was necessary to choose a particular use cycle to obtain a fair comparison among different stoves. In our study, we used the "water boiling test" procedure developed as a standard international method to compare energy efficiencies of different stoves (44). The water boiling test is a relatively short, simple simulation of a common cooking procedure in which a standard quantity of water is used to simulate food. The test includes high power and low power phases. The high power phase involves heating the standard quantity of water from the ambient temperature to boiling temperature as rapidly as possible. The low power phase follows in which the power is reduced to the lowest level needed to keep the water simmering. Except for coal burning, the burn cycles ranged from 35 to 60 min for all other types of fuel/stove combinations. Coal burning needs a longer cycle (up to 6 h). A pot

containing a known amount of water was placed on a tested stove during the entire burn cycle. Collected data included those necessary to determine thermal parameters such as burn rate and overall thermal efficiency of each fuel/stove combination. This procedure, thus, had added advantages of enabling the simultaneous measurement of emissions and energy efficiencies. The stove efficiency data were used for calculating emissions per standard cooking task.

The emissions from the cookstoves were collected using a sampling configuration which included, from upstream to downstream, a sampling probe, a filter holder, a pump, and a large Tedlar bag (80 L). For stoves having a flue, the probe was inserted in the flue. For those having no flue, there was a hood built above the stoves; and the probe was placed above the fire but under the hood. The flow rate of the sampling pump was adjusted to fill up the Tedlar bag throughout a burn cycle. After sampling, the filled Tedlar bag was tightly sealed before samples were taken out of the bag for analysis of CO<sub>2</sub>, CO, CH<sub>4</sub>, total non-methane organic compounds, carbonyl compounds, etc. Additional details of the sample collection and analysis can be found in our previous reports (41, 45, 46).

Among the three successful burn tests for each fuel/stove combination, two had the subsequent analyses for carbonyl compounds due to the limited budget. Results from several trial runs indicated that the background concentrations of carbonyl compounds did not vary significantly for different experimental runs. Thus, only one background air sample was taken for each fuel/stove combination. The background air sample had been taken indoors before a burn test was started. The background indoor concentrations were 2.7 ± 2.5 ppb for formaldehyde, 1.1 ± 1.2 ppb for acetaldehyde, and below detection limit for all the other measured carbonyl compounds (acetone, acrolein, propionaldehyde, crotonaldehyde, 2-butanone, isobutyraldehyde, butyraldehyde, isovaleraldehyde, valeraldehyde, hexaldehyde, benzaldehyde, *o*-tolualdehyde, *m,p*-tolualdehyde, and 2,4-dimethylbenzaldehyde).

**Measurement of Carbonyl Compounds.** The flue gas in a filled large Tedlar bag was drawn through a DNPH-coated cartridge (Waters Corp., USA) using a personal air sampling pump (SKC, Inc., USA). The Waters DNPH cartridge is a Sep-

Pak silica cartridge, coated with 2,4-dinitrophenylhydrazine (DNPH), and has been widely used to collect carbonyl compounds in various environments (23, 26, 47). Approximately 20 L of air was taken at a flow rate of  $\approx 1$  L/min. The exact sampling volume was recorded. The carbonyl-DNPH derivatives collected on the cartridges were extracted with 4 mL of HPLC-grade acetonitrile (ACN) and analyzed using an HPLC-UV technique which has been reported in detail in a previous study (23). The analytical detection limits ranged from 0.1 to 1 ng for various carbonyl species. The HPLC analytical conditions used were as follows: Nova-Pak C<sub>18</sub> column (3.9  $\times$  150 mm) and its guard column (Waters); gradient mobile phases: A = water/ACN/tetrahydrofuran 60/30/10 v/v/v, B = water/ACN 40/60 v/v, 100% A for 1 min, then linear gradient from 100% A to 100% B in 10 min, and then 100% B for another 10 min; mobile flow rate 1 mL/min; detector UV at 360 nm. Freshly synthesized DNPH-aldehyde derivatives (hydrazones) were used for identification and quantification. All calibration curves had linear regression coefficients  $R^2 > 0.99$  and near-zero intercepts.

**Determination of Emission Factors.** Carbon balance models have often been used to determine emission factors for large-scale open biomass fires (48–50). In a similar fashion, a carbon balance model has been used to determine emission factors for several cookstoves in a Manila pilot study (51) and for the 56 fuel/stove combinations in the full-scale study (41, 42). To utilize the carbon balance model, a complete carbon analysis is required. Thus, we have measured carbon mass in the fuel burnt, carbon mass in all emission products (CO<sub>2</sub>, CO, CH<sub>4</sub>, total non-methane organic compounds, and airborne particulate matter), carbon mass of the unburned fuel residues, and carbon mass in the char and ash generated by combustion. Methods and quality assurance procedures for all these measurements, along with descriptions of the model and necessary assumptions, can be found in detail in our previous reports (41, 45, 46). Using the model and the data from the complete carbon analysis, we have first calculated the CO<sub>2</sub> emission factor for each of the fuel/stove combinations tested (41). The emission factor of any other airborne species X (e.g., carbonyl compounds) can be calculated using the equation:

$$(\text{Ef})_X = (\text{Er})_X (\text{Ef})_{\text{CO}_2} \frac{(\text{MW})_X}{(\text{MW})_{\text{CO}_2}} \quad (1)$$

where  $(\text{Ef})_X$  and  $(\text{Ef})_{\text{CO}_2}$  are the emission factors of X and CO<sub>2</sub>, respectively, in grams per kilogram of fuel (or in grams per cooking task; see below);  $(\text{MW})_X$  and  $(\text{MW})_{\text{CO}_2}$  are the molecular weights of X and CO<sub>2</sub>, respectively; and  $(\text{Er})_X$  is the molar emission ratio of X to CO<sub>2</sub>, a dimensionless unit, and obtained from the equation:

$$(\text{Er})_X = \frac{C_{X,\text{emission}} - C_{X,\text{background}}}{C_{\text{CO}_2,\text{emission}} - C_{\text{CO}_2,\text{background}}} \quad (2)$$

where  $C_{X,\text{emission}}$  and  $C_{X,\text{background}}$  are the X concentration measured in the flue gas and in the background air, in ppm or any other molar concentration equivalent units, and  $C_{\text{CO}_2,\text{emission}}$  and  $C_{\text{CO}_2,\text{background}}$  are the CO<sub>2</sub> concentration measured in the corresponding flue gas and background air.

Since different amounts of fuels are needed for the same cooking task using different fuel/stove combinations, the task-based emission factor (mass of pollutant/cooking task),  $(\text{Ef})_{X,\text{g/task}}$ , rather than the fuel-mass-based task is a better performance index to compare the air pollution potential of different fuel/stove combinations (35, 38). The simplest task measure is the release per unit energy delivered to the cooking vessel (g/MJ). The following two parameters are needed to convert emissions per kilogram of fuel to emissions per task:

fuel calorific value ( $H$ , in MJ/kg) and stove efficiency ( $\eta$ , in %):

$$(\text{Ef})_{X,\text{g/task}} = \frac{(\text{Ef})_{X,\text{g/kg}}}{H\eta} \quad (3)$$

where  $J$  = energy delivered to the pot per standard task (MJ/task). In this paper, the standard cooking task was defined as one that would deliver 1 MJ heat to the pot (modified for simplification purposes from refs 35 and 52 where the cooking task was defined as one that would deliver 879 kJ heat).  $H$  and  $\eta$  were measured in the study and reported elsewhere (42). Once task-based CO<sub>2</sub> emission factors are obtained through eq 3, task-based emission factors for species X can be alternatively calculated using eq 1.

## Results

**Molar Emission Ratios.** Under a hypothetical condition for a perfectly complete combustion, all the carbon contained in the fuel would turn into CO<sub>2</sub>. Therefore, compounds other than CO<sub>2</sub> found in the flue gas are typically called products of incomplete combustion (PICs). The emission ratio of total PICs to CO<sub>2</sub> usually serves as an indicator of stove combustion efficiency (41); and the emission ratio of an individual PIC is often used as a relative parameter for the source strength of the PIC (51, 53). Emission ratios of the measured carbonyl compounds are presented in Table 2. The total carbonyl emission ratio is simply the summation of emission ratios of all the identified carbonyls. In Table 2, a blank cell indicates that the net carbonyl concentration was equal to or less than zero, showing that the corresponding burn did not generate a detectable amount of the carbonyl compound.

The results show that formaldehyde and acetaldehyde concentrations in the flue gases were higher than the background levels for the tested stoves. Burning of biomass fuels (WR, MR, FW, and BW; see Table 1 for the fuel definitions), in the vast majority of the burn tests, appeared to produce additional carbonyl compounds such as acetone, acrolein, propionaldehyde, crotonaldehyde, 2-butanone, isobutyraldehyde, butyraldehyde, isovaleraldehyde, valeraldehyde, hexaldehyde, benzaldehyde, *o*-tolualdehyde, *m,p*-tolualdehyde, and 2,4-dimethylbenzaldehyde. (The sum of *m*-tolualdehyde and *p*-tolualdehyde was reported because they could not be separated by the analytical method.) Although most of the above-listed carbonyls were identified in the flue gases of kerosene (KE) and LPG stoves, these carbonyls were rarely generated by burning coal (HC, CB, and UC), coal gas (CG), and natural gas (NG).

As shown in Table 2, the difference between the results derived from the two separate tests for each fuel/stove combination, in some cases, is large. This may result mainly from relatively large variations in combustion conditions (e.g., natural variability due to differences in fuel burn rate and in operator behavior) between the two tests (which are difficult to control especially for the solid fuels). Based on these measurements, nevertheless, ranges of emission ratios of the most abundant carbonyl compounds were as follows: formaldehyde,  $36 \times 10^{-6}$  to  $280 \times 10^{-6}$  for biomass burning and  $2.7 \times 10^{-6}$  to  $83 \times 10^{-6}$  for fossil fuel burning; acetaldehyde,  $26.4 \times 10^{-6}$  to  $265 \times 10^{-6}$  for biomass burning and  $0.7 \times 10^{-6}$  to  $104 \times 10^{-6}$  for fossil fuel burning; and total carbonyl,  $77.4 \times 10^{-6}$  to  $1032 \times 10^{-6}$  for biomass burning and  $3.8 \times 10^{-6}$  to  $263 \times 10^{-6}$  for fossil fuel burning.

**Emission Factors.** The emission factors of the 17 carbonyl compounds, along with CO<sub>2</sub> emission factors, are shown in Tables 3 and 4 on a fuel-mass basis and the defined cooking-task basis, respectively, for each of the 22 fuel/stove combinations. These emission factors were derived from the

TABLE 2. Molar Emission Ratios to CO<sub>2</sub> of Carbonyls Identified in Flue Gas (Dimensionless Unit  $\times 10^{-6}$ )<sup>a</sup>

fuel/ stove/ flue	formal- dehyde	acetal- dehyde	ace- tone	acro- lein	propional- dehyde	crotonal- dehyde	2-buta- none	isobutyr- aldehyde	butyr- aldehyde	benzal- dehyde	isovaleral- dehyde	valeral- dehyde	<i>o</i> -tolual- dehyde	<i>m,p</i> -tolual- dehyde	hexal- dehyde	2,4-dimethyl- benzal- dehyde	total
WR/brick/1	80.7	46.3	1.8	7.5	4.7	2.9	7.3	4.1	3.0	6.2	3.9			1.0		0.7	170
	65.4	44.5			5.6	2.3	3.2		6.7	5.0	4.2			0.9			138
WR/imp/1	146	168	9.4	128	27.3	14.1	68.8	21.6	5.0	11.8	6.8	0.8	0.9	3.4		1.5	613
	225	198	9.8	271	51.3	26.4	133	58.3	16.1	11.2		15.7	2.8	10.9		2.9	1033
MR/brick/1	36.0	36.1	3.1		3.6	2.1				1.4	1.3			1.7			85.4
	55.8	37.8			2.6		4.4		5.1	3.7							109
MR/imp/1	112	67.8	13.4	4.3	5.3	9.8			5.4	8.0	0.4		1.7	2.8	1.6		232
	123	32.4	7.3	9.6	28.5	19.8	67.0	22.8	7.3	20.0		6.0	2.1	9.9		1.7	357
FW/brick/1	40.5	26.4	2.1	1.0	1.6	2.5	1.4			1.2	0.8						77.4
	40.5	45.5	2.4		2.4	5.0	8.0			1.5		1.5		3.1			110
FW/imp/1	148	45.3	38.8	15.5	36.3	36.1	36.0	6.6	17.9	11.6		3.9	6.6	33.4	8.2	2.4	446
	81.3	44.5	10.3		3.9	5.9	4.2	1.0	2.9	8.4		2.4	0.6	2.7	0.5	0.7	169
FW/Indian/0	281	128	26.5	8.0	21.5	17.4	38.6			14.2		3.4	2.8	12.9	2.1	3.0	559
	132	71.0	14.2	4.0	6.6	10.9	4.7	1.8	7.9	4.9	1.3		1.1	4.6	1.3		267
BW/brick/1	104	99.7	11.0		7.2	11.9	5.2	6.1	13.2	2.1		2.0		5.0			268
	92.4	91.4	16.4	3.3	10.0	14.1	5.6	3.2	8.4	2.5	1.2	2.1	1.3	4.6	1.7		258
BW/Indian/0	267	175	22.4		17.2	22.2	5.9	8.4	8.4	16.1		9.4	2.2	6.0	1.3	1.8	555
	225	265	38.5	9.8	24.7	21.2	26.8	6.4	13.4	21.1	6.4		3.6	13.1	3.4	2.1	681
HC/metal/1	5.1	4.2										0.6			0.7		10.6
	8.7	5.3															13.9
HC/metal/0	4.9	3.8									0.6						9.3
	5.1	15.9															21.0
HC/imp/0	11.5	10.9															22.4
	28.1	1.9	0.2		0.8												31.0
CB/metal/1	2.7	0.7												0.4			3.8
	4.7	4.1															8.9
CB/metal/0	42.6	8.1	0.0														50.8
	16.4	9.7	3.8														29.9
UC/brick/1	40.9	44.7	3.5		4.5	5.6	2.4										102
	19.0	24.5															43.5
KE/wick/0	34.8	45.6	5.2	1.9	10.4	5.7	3.1	7.3	4.6	0.9		3.4	0.7	2.6	0.9	0.6	128
	16.9	14.1		5.4	2.1	3.0	5.7		5.3	1.4	1.4		1.5				56.9
KE/pres/0	41.8	15.4				10.6				4.4		5.5					77.7
	83.0	34.8	0.7		7.5	5.7		3.7	4.7	2.0	1.6			3.1			147
LPG/trad/0	55.7	29.2			8.6	6.3				9.2	3.5			2.0			114
	51.7	20.6			13.2	17.0				17.0	1.7			5.9			127
LPG/inf/0	73.2	104	9.1	8.9	11.7	18.1		5.6	2.1	0.5		0.8	3.7	21.1	2.3	1.5	263
	44.4	62.6	4.3	3.3	4.7	7.6	1.4		2.2	0.8	0.6		1.3	8.6			142
CG/trad/0	13.8	6.3				1.2				0.3		0.6					22.1
	31.0	9.6															40.6
NG/trad/0	30.3	20.6	1.2		1.7	1.5				0.8	0.7			1.2			58.0
	29.0	13.1			3.2					3.2					2.2		50.6
NG/inf/0	19.0	5.8			2.1	1.6				0.7	0.5			0.8			30.5
	34.6	3.2				4.0				3.5							45.2

<sup>a</sup> A blank cell indicates that the flue gas concentration was not greater than the background concentration.



TABLE 3. Emission Factors of Carbonyl Compounds, mg/kg, for the 22 Fuel/Stove Combinations Tested<sup>a</sup>

fuel/ stove/ flue	CO <sub>2</sub> (g/kg)	formal- dehyde	acetal- dehyde	ace- tone	acrol- ein	propional- dehyde	crotonal- dehyde	2-buta- none	isobutyr- aldehyde	butyr- aldehyde	benzal- dehyde	isovaleral- dehyde	valeral- dehyde	<i>o</i> -tolual- dehyde	<i>m,p</i> -tolual- dehyde	hexal- dehyde	2,4-dimethyl- benzal- dehyde	total
WR/brick/1	1342	73.8	62.2	3.1	12.8	8.2	6.2	16.0	8.9	6.5	20.0	10.2					2.9	234
	1368	61.0	60.8					7.2		15.0	16.5	11.3						190
WR/imp/1	1162	116	195	14.5	189.0	41.9	26.2	131.0	41.0	9.6	33.0	15.4	1.8	3.0			5.3	830
	832	128	165	10.8	287.7	56.4	35.0	181.9	79.5	21.9	22.5		25.6	6.4	34.7		7.4	1062
MR/brick/1	1166	28.6	42.1	4.8		5.6	3.8				3.9	2.9			5.5			97
	1184	45.1	44.7			4.0		8.5		9.8	10.5							123
MR/imp/1	1155	88.1	78.3	20.4	6.3	8.1	18.1			10.1	22.3	1.0		5.5	7.8	4.1		270
	1019	85.4	33.0	9.9	12.5	38.3	32.2	111.9	38.1	12.2	49.2		12.0	5.8	41.6		5.4	487
FW/brick/1	1540	42.6	40.7	4.3	1.9	3.2	6.1	3.6			4.3	2.3						109
	1522	42.0	69.2	4.8		4.8	12.1	19.9			5.4		4.6		13.3			176
FW/imp/1	1591	160	72.2	81.4	31.5	76.2	91.6	94.0	17.2	46.6	44.6		12.0	28.6	146	29.8	11.4	943
	1595	88.4	71.0	21.7		8.1	15.0	11.1	2.6	7.6	32.5		7.6	2.7	7.4	1.9	3.2	281
FW/Indian/0	1005	192	129	35.2	10.2	28.6	27.9	63.6			34.3		6.7	7.6	56.1	4.8	9.1	605
	1588	143	113	29.8	8.0	13.8	27.6	12.2	4.7	20.4	18.7	4.1		4.8	19.1	4.7		424
BW/brick/1	1522	108	152	22.1		14.5	28.9	12.9	15.2	33.0	7.7		5.9		21			421
	1525	96.1	139	33.0	6.5	20.2	34.3	13.9	7.9	21.0	9.2	3.5	6.2	5.3	18	5.8		420
BW/Indian/0	1434	261	251	42.4		32.5	32.5	52.1	13.9	19.7	55.6		26.3	8.6	23	4.2	7.8	799
	1397	215	371	71.1	17.4	45.5	47.2	61.4	14.7	30.6	71.1	17.4		13.9	72.8	10.7	8.9	1068
HC/metal/1	2042	7.0	8.6										2.2			3.4		21
	2094	12.4	11.0															23
HC/metal/0	2207	7.3	8.4									2.8						18
	1936	6.7	30.8															38
HC/imp/0	2134	16.7	23.3															40
	2163	41.4	4.1	0.7		2.3												49
CB/metal/1	1151	2.2	0.8												1.2			4
	1206	3.9	5.0															9
CB/metal/0	1272	37.0	10.3															47
	1426	16.0	13.8	7.1														37
UC/brick/1	1811	50.5	80.9	8.5		10.8	16.1	7.0										174
	1591	20.6	39.0															60
KE/wick/0	3124	74.2	143	21.6	7.5	43.1	28.4	15.9	37.4	23.4	7.1		21.0	5.5	22.0	6.3	5.8	462
	3120	36.0	44.1		21.4	8.5	15.1	29.0		27.2	10.7	8.7		13.1				214
KE/pres/0	3124	89.1	48.0				52.8				33.4		33.5					257
	3135	177	109	2.8		31.2	28.3		19.0	23.9	15.1	9.8			26.0			443
LPG/trad/0	3084	117	89.9			35.1	30.8				68.3	21.0			16.8			379
	3080	109	63.4			53.6	83.6				126	10.2			48.9			494
LPG/inf/0	3062	153	320	36.9	34.7	47.2	88.3		28.1	10.6	3.4		4.7	31.0	177	16.0	13.5	964
	3073	93.1	192	17.3	12.8	18.9	37.3	7.0		11.1	6.1	3.3		10.6	43.5			453
CG/trad/0	1844	17.3	11.6				3.4				1.1		2.2					36
	1844	38.9	17.7															57
NG/trad/0	3443	71.2	71.1	5.5		7.6	8.0				7.0	5.0			10.9			186
	3443	68.0	45.1			14.4					26.4					17.3		171
NG/inf/0	3443	44.5	20.1			9.6	8.8				5.6	3.5			7.3			99
	3443	81.1	10.9				22.2				28.8							143

<sup>a</sup> A blank cell indicates that the flue gas concentration was not greater than the background concentration.

TABLE 4. Emission Factors of Carbonyl Compounds, mg/(Task of 1 MJ Delivered), for the 22 Fuel/Stove Combinations Tested<sup>a</sup>

fuel/ stove/ flue	CO <sub>2</sub> (g/kg)	formal- dehyde	acetal- dehyde	ace- tone	acro- lein	propional- dehyde	crotonal- dehyde	2-buta- none	isobutyr- aldehyde	butyr- aldehyde	benzal- dehyde	isovaleral- dehyde	valeral- dehyde	o-tolual- dehyde	m,p-tolual- dehyde	hexal- dehyde	2,4-dimethyl- benzal- dehyde	total
WR/brick/1	969	53.3	44.9	2.3	9.3	6.0	4.4	11.6	6.4	4.7	14.4	7.4			2.4		2.1	169
	909	40.5	40.4			6.7	3.3	4.8		9.9	11.0	7.5			1.3			126
WR/imp/1	552	55	93	6.9	89.7	19.9	12.4	62.2	19.5	4.5	15.7	7.3	0.9	1.4	3.3		2.5	394
	353	54	70	4.6	121.9	23.9	14.8	77.1	33.7	9.3	9.5		10.9	2.7	17.7		3.1	453
MR/brick/1	595	14.6	21.5	2.5		2.9	2.0				2.0	1.5			2.9			50
	615	23.4	23.2			2.1		4.4		5.1	5.4							64
MR/imp/1	372	28.4	25.2	6.6	2.0	2.6	5.8			3.3	7.2	0.3		1.8	2.7	1.3		87
	361	30.2	11.7	3.5	4.4	13.6	11.4	39.6	13.5	4.3	17.4		4.2	2.1	17.3		1.9	175
FW/brick/1	639	17.7	16.9	1.8	0.8	1.3	2.5	1.5			1.8	1.0						45
	919	25.4	41.8	2.9		2.9	7.3	12.0			3.3		2.8		3.4			102
FW/imp/1	407	41	18.5	20.8	8.1	19.5	23.5	24.1	4.4	11.9	11.4		3.1	7.3	37	7.6	2.9	241
	408	22.6	18.2	5.5		2.1	3.9	2.8	0.7	1.9	8.3		2.0	0.7	1.9	0.5	0.8	72
FW/Indian/0	257	49	33	9.0	2.6	7.3	7.1	16.3			8.8		1.7	1.9	12.5	1.2	2.3	153
	353	32	25	6.6	1.8	3.1	6.1	2.7	1.0	4.5	4.2	0.9		1.1	8.4	1.0		98
BW/brick/1	666	47	66	9.7		6.4	12.6	5.6	6.6	14.4	3.4		2.6		10			185
	711	44.8	65	15.4	3.0	9.4	16.0	6.5	3.7	9.8	4.3	1.6	2.9	2.5	7	2.7		194
BW/Indian/0	548	100	96	16.2		12.4	19.9	19.9	5.3	7.5	21.2		10.1	3.3	9	1.6	3.0	306
	547	84	145	27.8	6.8	17.8	18.5	24.0	5.8	12.0	27.9	6.8		5.4	22.1	4.2	3.5	412
HC/metal/1	619	2.1	2.6										0.7			1.0		6.4
	677	4.0	3.6															7.6
HC/metal/0	757	2.5	2.9									0.9						6.3
	453	1.6	7.2															8.8
HC/imp/0	237	1.9	2.6															4.4
	233	4.5	0.4	0.1		0.2												5.2
CB/metal/1	462	0.9	0.3												0.3			1.5
	325	1.0	1.3															2.4
CB/metal/0	251	7.3	2.0															9.4
	282	3.2	2.7	1.4														7.3
UC/brick/1	440	12.3	19.7	2.1		2.6	3.9	1.7										42
	229	3.0	5.6															8.6
KE/wick/0	157	3.7	7	1.1	0.4	2.2	1.4	0.8	1.9	1.2	0.4		1.1	0.3	1.1	0.3	0.3	23
	152	1.8	2.2		1.0	0.4	0.7	1.4		1.3	0.5	0.4		0.6				10
KE/pres/0	146	4.2	2.2				2.5				1.6		1.6					12
	164	9	6	0.1		1.6	1.5		1.0	1.3	0.8	0.5			1.2			23
LPG/trad/0	147	6	4.3			1.7	1.5				3.2	1.0			0.7			18
	132	5	2.7			2.3	3.6				5	0.4			2.1			21
LPG/inf/0	133	7	14	1.6	1.5	2.1	3.8		1.2	0.5	0.1		0.2	1.3	8	0.7	0.6	42
	131	4.0	8	0.7	0.5	0.8	1.6	0.3		0.5	0.3	0.1		0.5	2.2			20
CG/trad/0	94	0.9	0.6				0.2				0.1		0.1					1.8
	83	1.8	0.8															2.5
NG/trad/0	123	2.5	2.5	0.2		0.3	0.3				0.2	0.2			0.4			6.6
	124	2.5	1.6			0.5					1.0					0.6		6.2
NG/inf/0	110	1.4	0.6			0.3	0.3				0.2	0.1			0.2			3.2
	109	2.6	0.3				0.7				0.9							4.5

<sup>a</sup> A blank cell indicates that the flue gas concentration was not greater than the background concentration.

TABLE 5. Carbonyl Emission Factors (mg/kg): Summary Statistics by Fuel Types<sup>a,b</sup>

	crop residue (n = 8)		wood (n = 10)		coal (n = 12)		kerosene (n = 4)		LPG (n = 4)		coal gas (n = 2)		natural gas (n = 4)	
	mean	c.v.	mean	c.v.	mean	c.v.	mean	c.v.	mean	c.v.	mean	c.v.	mean	c.v.
formaldehyde	78.3	0.43	135	0.54	18.5	0.87	94.3	0.63	118	0.21	28.3	0.54	66.3	0.23
acetaldehyde	85.1	0.71	141	0.71	19.7	1.14	85.8	0.56	166	0.70	14.7	0.29	36.7	0.73
acetone	10.6	0.60	34.6	0.73	5.4	0.77	12.2	1.09	27.1	0.51			5.5	
acrolein	101	1.27	12.6	0.84			14.4	0.68	23.8	0.65				
propionaldehyde	21.6	0.95	24.7	0.91	6.5	0.92	27.6	0.64	38.7	0.39			10.5	0.33
crotonaldehyde	18.1	0.74	32.3	0.79	16.1		31.1	0.50	60.2	0.50	3.4		13.0	0.61
2-butanone	59.3	1.29	33.2	0.89	7.0		32.0	0.41	7.0					
isobutyraldehyde	41.9	0.69	10.9	0.53			28.2	0.46	28.1					
butyraldehyde	12.1	0.41	25.6	0.49			24.8	0.08	10.9	0.04				
benzaldehyde	22.2	0.63	28.3	0.82			16.6	0.71	51.0	1.14	1.1		16.9	0.73
isovaleraldehyde	8.2	0.74	6.8	1.04	2.8		9.3	0.08	11.5	0.77			4.2	0.25
valeraldehyde	13.1	0.91	9.9	0.77	2.2		27.3	0.32	4.7		2.2			
o-tolualdehyde	5.2	0.29	10.2	0.87			9.3	0.57	20.8	0.70				
m,p-tolualdehyde	14.8	1.09	41.8	1.06	1.2		24.2	0.13	71.6	0.88			9.1	0.28
hexaldehyde	4.1		8.8	1.09	3.4		6.3		16.0				17.3	
2,4-dimethylbenzaldehyde	5.3	0.35	8.1	0.37			5.8		13.5					
total carbonyls <sup>b</sup>	399	0.89	525	0.62	43.3	1.02	344	0.37	573	0.46	46.2	0.33	150	0.25

<sup>a</sup> n = sample number, c.v. = coefficient of variation = (standard deviation)/mean. <sup>b</sup> Derived from Table 3.

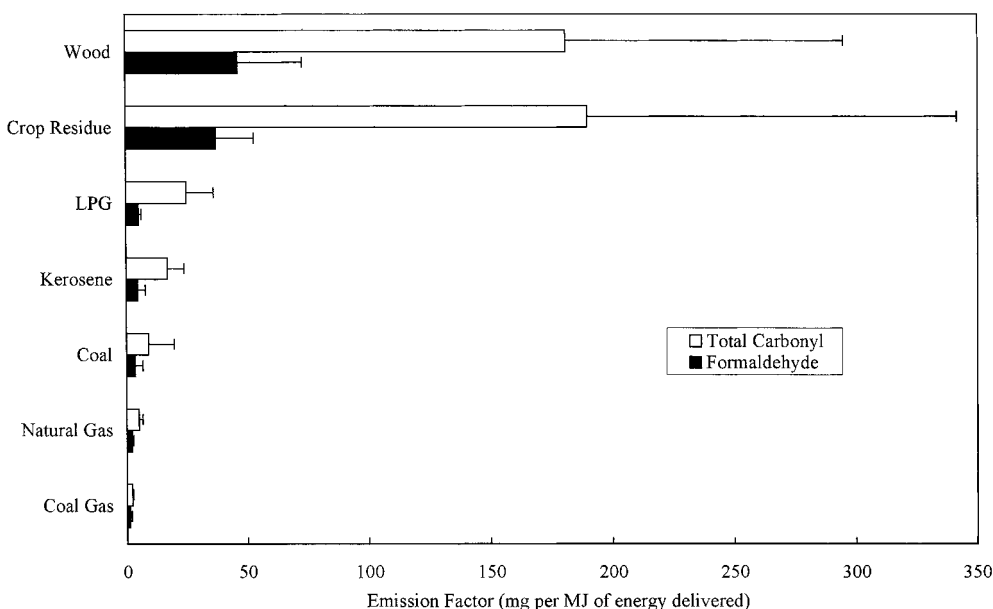


FIGURE 1. Task-based mean emission factors for the seven types of fuel tested (error bar represents 1 SD).

values of the emission ratios listed in Table 2 and the CO<sub>2</sub> emission factors using eq 1.

### Discussion

**Fuel Types and Carbonyl Emissions.** The 22 fuel/stove combinations tested for carbonyl emissions covered 7 fuel types: crop residue, wood, coal, kerosene, LPG, coal gas, and natural gas. Within these fuel types, crop residue includes WR and MR; wood included FW and BW; coal included HC, CB, and UC. The fuel-mass-based emission factors, summarized by fuel type in Table 5, have a wide range. Besides the biomass fuels, LPG and kerosene also have substantially higher emission factors compared to the other types of fossil fuels.

Although burning the same amount (mass) of kerosene, LPG, and the biomass fuels emits a comparable amount of carbonyl compounds, performing the same cooking task using the biomass fuels would produce significantly higher carbonyl emissions than using LPG or kerosene due to higher energy contents in LPG and kerosene and higher energy efficiencies of LPG and kerosene stoves (see Figure 1). The results, as shown in Figure 1, also indicate that task-based

emission factors of formaldehyde and the total carbonyls were different among the five types of fossil fuels tested, ranking as follows: LPG > kerosene > coal > natural gas > coal gas. It is interesting to notice that coal seems to be a cleaner fuel than LPG and kerosene if only concerning the emissions of carbonyl compounds. Burning coal in cookstoves, however, may generate higher emissions of other PICs such as particulate matter and CO than other types of fossil fuels (36, 54).

**Stove Types and Carbonyl Emissions.** The results show that the carbonyl emissions were dependent not only on the fuel type but also on the stove type. As shown in Figure 2, for example, performing the same cooking task using the same types of fuel but different types of stoves produced different formaldehyde emissions. Interestingly, the improved stoves seemed to have higher formaldehyde emission factors: WR/imp/1 > WR/brick/1; MR/imp/1 > MR/brick/1; FW/imp/1 > FW/brick/1. This can be explained as follows: An improved stove is named so generally based on the fact that the stove has a higher overall energy efficiency. The overall efficiency is a product of combustion efficiency and heat transfer efficiency (44, 54). An increase in heat transfer

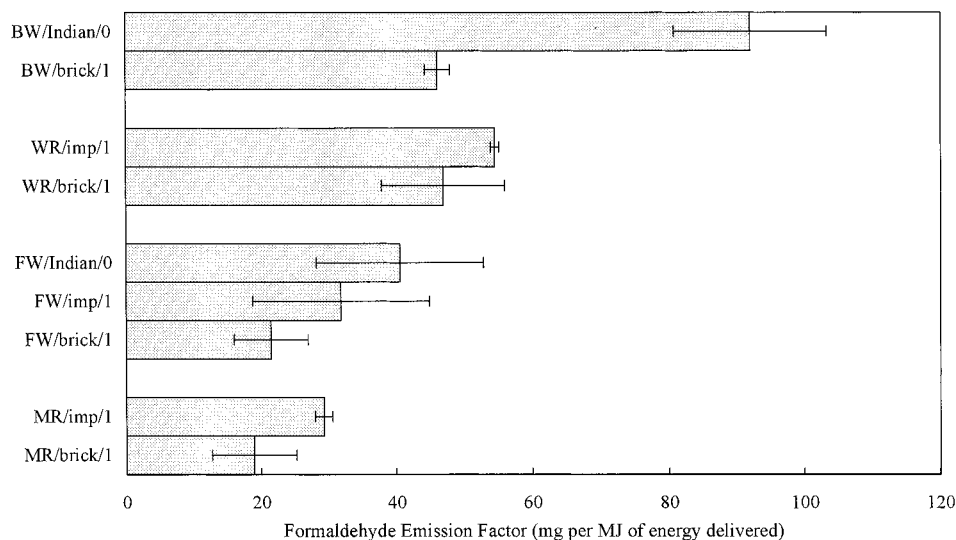


FIGURE 2. Formaldehyde emission factors as a function of stove type (error bar represents 1 SD).

efficiency, by better insulating the stove, usually leads to a decrease in combustion efficiency due to the reduction of air supply to the fire (35). Consequently, the decreased *combustion efficiency* would turn fuel carbon into more PICs such as carbonyl compounds. This implies that the cookstove improvement, if only considering overall energy efficiency, may cause higher air pollution potentials.

**Potential Exposures to Carbonyl Emissions.** To estimate the significance of the contribution a cookstove can make on the exposure to the carbonyl compounds, we use the following single-compartment mass balance model to estimate the indoor formaldehyde concentration  $C$  ( $\text{g}/\text{m}^3$ ) as a function of time  $t$  (hr):

$$\frac{dC}{dt} = \frac{F(\text{Ef})_{\text{HCHO}}}{V} - kC \quad (4)$$

where  $F$  = fuel burn rate ( $\text{kg}/\text{h}$ );  $(\text{Ef})_{\text{HCHO}}$  = formaldehyde emission factor ( $\text{g}/\text{kg}$ );  $V$  = volume of house ( $\text{m}^3$ );  $k$  = air exchange rate ( $\text{h}^{-1}$ ). Assuming the initial formaldehyde concentration (before the stove was lighted) is zero, the formaldehyde concentration ( $C_t$ ) in room air at time  $t$  during the combustion is

$$C_t = \frac{F(\text{Ef})_{\text{HCHO}}}{Vk} (1 - e^{-kt}) \quad (5)$$

Equation 5 is derived based upon the following assumptions: (1) There are no other formaldehyde sources than the cookstove emissions (other formaldehyde sources are most likely to be insignificant for village houses); (2) there are no other formaldehyde sinks than ventilation (other sinks such as dry deposition would be negligible for village houses which typically have high air exchange rates); (3) the room air is well mixed (see below); (4)  $(\text{Ef})_{\text{HCHO}}$  is constant during the course of combustion (here we use the time-averaged formaldehyde emission factors); (5) the fuel burn rate,  $F$ , is constant; and (6) all the formaldehyde emitted is directly released into the room (which is the case when the fuel is burnt in a cookstove without use of any flue, hood, or other venting devices). Using these assumptions, we have calculated, for illustrative purposes, the formaldehyde concentration in a typical  $40 \text{ m}^3$  village house where the fuel wood (FW) is burnt in the India-made metal stove (fuel/stove # 7 in Table 1). After the fuel is continuously burnt for 1 h at a typical burn rate of  $1.67 \text{ kg}/\text{h}$  to cook a meal, the peak formaldehyde concentration, i.e.,  $C_{t=1\text{h}}$ , in the house would be  $370 \mu\text{g}/\text{m}^3$  (0.30 ppm) if  $k = 19 \text{ h}^{-1}$  (high end of the  $k$  range

for village houses) and  $3030 \mu\text{g}/\text{m}^3$  (2.5 ppm) if  $k = 2 \text{ h}^{-1}$  (low end of the  $k$  range for village houses) (38, 56). The time-averaged formaldehyde concentration,  $\bar{C}$ , during the 1 h combustion can be calculated from the equation:

$$\bar{C} = \frac{\int_0^T C_t dt}{T} = \frac{F(\text{Ef})_{\text{HCHO}}}{Vk} \left[ 1 + \frac{1}{kT} (e^{-kT} - 1) \right] \quad (6)$$

where  $T$  = combustion duration = 1 h. When  $k = 19 \text{ hr}^{-1}$ ,  $\bar{C} = 350 \mu\text{g}/\text{m}^3$  (0.29 ppm), and when  $k = 2 \text{ h}^{-1}$ ,  $\bar{C} = 1990 \mu\text{g}/\text{m}^3$  (1.6 ppm).

The above model-estimated  $\bar{C}$  range, corresponding to a wide range of air exchange rate, is comparable to the time-averaged formaldehyde concentrations derived from previous integrated measurements. For example, Raiyani and co-workers measured indoor formaldehyde levels during cooking hours in houses located in the eastern peripheral area of the Ahmedabad agglomeration, India (39). They have reported that arithmetic means and geometric means of indoor formaldehyde concentrations for 20 houses using wood cookstoves were  $916 \mu\text{g}/\text{m}^3$  and  $652 \mu\text{g}/\text{m}^3$ , respectively. Another study, however, reported a substantially lower indoor formaldehyde concentration range which was  $145\text{--}182 \mu\text{g}/\text{m}^3$  after 1 h of fuelwood combustion in a stove without flue (57). The difference between these reported indoor formaldehyde concentrations is due to large differences in ventilation rate, house volume, and stove design.

Using the above model, we were able to estimate the peak formaldehyde concentration which could not be obtained through time-integrated measurements. The peak concentrations, however, may be important to know, because some acute health effects may mainly result from exposures to high peak concentrations rather than accumulative exposures to relatively low concentrations. It is reported that 0.5–1 ppm formaldehyde is detectable by odor, 2–3 ppm produces mild irritation, and 4–5 ppm is intolerable to most people (2). The peak formaldehyde concentrations resulting from cookstove biomass combustion, therefore, may be high enough to cause some or all of these acute health effects, depending upon house parameters (e.g.,  $k$  and  $V$ ) and individuals' susceptibility. Given that houses are not necessarily well-mixed and that a cook may stay very close to the fire, formaldehyde concentrations to which the cook is actually exposed may be even higher than those estimated using the model. In addition, other carbonyl compounds in the cookstove emission could add extra health risks. For example, acrolein, more irritating than formaldehyde (2),



was identified in the flue gases of most biomass cookstoves tested in this study.

**Summary and Implications.** In this paper, we present a new database of emission factors of 17 carbonyl compounds for 22 fuel/stove combinations in common use in developing countries. Cookstoves are individually small, but so numerous that they could have significant influences on carbonyl emission inventories. The results from this study indicate that the emission factors were not only a function of the fuel type, but also of the stove type, implicating that using this new database would be able to provide more accurate estimates of carbonyl emissions from domestic cookstoves than the emission factors normally measured under open or large-scale combustion conditions (e.g., open fire of biomass, industrial scale burning of fossil fuels).

In general, because the household sources emit directly into the places and at the times of human occupancy, exposures to certain air pollutants derived from these small sources are often greater than those derived from large outdoor sources (58, 59). This is very likely to be true for carbonyl exposures occurring in the village houses where biomass cookstoves are used. Our model-estimated indoor concentration ranges of formaldehyde during cooking hours, along with those measured by previous investigators, can be high enough to cause certain acute health effects, depending upon house characteristics (e.g., volume, air exchange rate) and individuals' susceptibility. Compared to typical indoor formaldehyde levels of 5–50 ppb found in urban homes where the major sources of formaldehyde are building materials and furniture (4, 19, 23), the village house levels of formaldehyde resulting from biomass combustion are much higher. Using open-fire biomass cookstoves certainly results in high exposures to formaldehyde and other carbonyl compounds.

Among the seven fuel types tested, the emission factors ranged widely, suggesting that switching usage of a dirtier fuel to a cleaner one would reduce the emissions and consequently the exposure. For example, if a natural gas stove is used instead of the Indian wood stove in the village house described above, 1 h continuous burning (at a typical gas range burn rate = 0.2 kg/h) would generate a peak indoor formaldehyde concentration of only  $\approx 0.01$  ppm (at  $k = 19$  h<sup>-1</sup>) and  $\approx 0.1$  ppm (at  $k = 2$  hr<sup>-1</sup>). Unfortunately, this strategy is not very practical in many rural areas in developing countries because wood and other biomass are still the major fuels for domestic cooking/heating and fossil fuel supplies are expensive and often unreliable. A simple but relatively effective way to reduce the exposure is to use cookstoves with flues, chimneys, or hoods instead of releasing pollutants directly into the kitchen (40). For example, the formaldehyde concentration when using a stove with flue was about half of that when using a flueless stove in the same indoor environment measured by Kandpal and co-workers (57). However, the emissions to the atmosphere would be the same unless the thermal performance of the stove is changed with the addition of flue, chimney, or hood.

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