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Mutagenicity- and Pollutant-Emission Factors of Solid-Fuel Cookstoves: Comparison to Other Combustion Sources

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Abstract

Background: Emissions from solid fuels used for cooking cause ~4 million premature deaths per year. Advanced solid-fuel cookstoves are a potential solution, but they should be assessed by appropriate performance indicators, including biological effects.

Objective: We evaluated two categories of solid-fuel cookstoves for 8 pollutant- and 4 mutagenicity-emission factors, correlated the mutagenicity-emission factors, and compared them to those of other combustion emissions.

Methods: We burned red oak in a 3-stone fire (TSF), a natural-draft stove (NDS), and a forced-draft stove (FDS); we combusted propane as a liquified petroleum gas control fuel. We determined emission factors based on useful energy (megajoules delivered, MJ_d) for carbon monoxide, nitrogen oxides (NO_x), black carbon, methane, total hydrocarbons, 32 polycyclic aromatic hydrocarbons, PM_{2.5}, levoglucosan (a wood-smoke marker), and mutagenicity in *Salmonella*.

Results: Other than NO_x the emission factors per MJ_d correlated highly among each other ($r^2 \geq 0.92$); NO_x correlated 0.58-0.76 with the other emission factors. Excluding NO_x, the NDS and FDS reduced the emission factors on average 68 and 92%, respectively, relative to the TSF. Nonetheless, the mutagenicity-emission factor based on fuel energy used (MJ_{thermal}) for the most efficient stove (FDS) was intermediate to that of a large diesel bus engine and a small diesel generator.

Conclusions: Both mutagenicity- and pollutant-emission factors may be informative for characterizing cookstove performance. However, mutagenicity-emission factors may be especially useful for characterizing potential health effects, and should be evaluated in relation to health outcomes in future research. A FDS operated as intended by the manufacturer is safer than a TSF, but without adequate ventilation, it still results in poor indoor air quality.

Introduction

Although humans may have harnessed the power of fire as long as 1 million years ago (Berna et al. 2012), approximately 40% of us still cook and heat with fire in ways almost indistinguishable from those of our distant ancestors (IARC 2010). Biomass in the form of various solid fuels (wood, straw, dung, charcoal, biomass briquettes, etc.) has yet to be replaced by modern energy carriers such as electricity and natural/petroleum gas for an estimated 2.8 billion people (Bonjour et al. 2013). In the U.S. 500,000 to 600,000 low-income people are estimated to be exposed to hazardous emissions from burning solid fuels inside their homes (Rogalsky et al. 2014). Smith et al. (2014) concluded that in 2010 household air pollution was responsible for 3.9 million premature deaths and ~4.8% of lost healthy life years, making it the most important environmental risk factor globally.

These deaths are due to a variety of diseases, including cardiovascular (Smith and Peel 2010), chronic obstructive pulmonary disease (Kurmi et al. 2010), low birth weight and stillbirth (Pope et al. 2010), upper-respiratory infections in children (Dherani et al. 2008), and lung cancer (IARC 2010). Household air pollution affects women disproportionately, being the second highest risk factor in terms of the global burden of disease for women (Lim et al. 2012).

Global efforts are underway to deliver electricity and gas to underserved populations to replace solid-fuel cookstoves (Pachauri et al. 2013) because this is the ultimate step to improve health outcomes (Balakrishnan et al. 2014; Pachauri et al. 2013; Smith 2014). An interim approach involves introducing solid-fuel cookstoves with new designs that are safer than the ones in use currently. Thus, the World Health Organization (WHO 2014) has developed air-quality guidelines by which cookstoves can be evaluated, and the International Organization for Standardization (ISO 2012) established an International Workshop Agreement to provide interim

guidelines for evaluating cookstoves on four performance indicators: (1) fuel efficiency, (2) total emissions, (3) indoor emissions, and (4) safety (from burns and injuries). The indoor-emissions indicator is related to WHO guidelines for air quality, but a specific performance indicator was not included for health effects.

Approximately 20 studies (Jetter et al. 2012; Just et al. 2013; Preble et al. 2014) have assessed many cookstoves burning a variety of solid fuels in the field and laboratory for multiple performance measures. Based on an analysis of 22 cookstoves, Jetter et al. (2012) suggested some metrics that could be used for setting international standards for ranking cookstove performance. These included emissions of carbon monoxide (CO) and particulate matter with an aerodynamic diameter $\leq 2.5 \mu\text{m}$ (PM_{2.5}) based on useful energy delivered to the contents of the cooking vessel.

However, an important question is whether these indicator pollutants provide an adequate indication of the health effects of the stoves. To help address this issue, we have evaluated two cookstoves representing two general categories of solid-fuel cookstoves: a natural-draft stove (NDS) and a forced-draft stove (FDS) (Figure 1). The rocket-type NDS is a vertical tube with an opening at the bottom to introduce the fuel, permitting air to flow by natural convection through the combustion chamber. In contrast, the FDS contains a fan in its base that forces air into the combustion chamber. We burned low-moisture red oak of the size prescribed by the manufacturers of these stoves under laboratory conditions similar to Jetter et al. (2012) and in a 3-stone fire (TSF) for comparison. We also characterized the emissions from a propane stove as an example of a clean-burning liquified petroleum (LP) gas fuel.

We extended the assessments of Jetter et al. (2012) to also include black carbon (BC), and we also evaluated dichloromethane (DCM) extracts of the PM_{2.5} for the concentrations of 32

polycyclic aromatic hydrocarbons (PAHs) and levoglucosan (a marker of wood smoke), as well as for mutagenicity in *Salmonella*. We expressed all results as emission factors, correlated them, determined the percent reduction of the emission factors by the two stoves relative to those of a TSF, and compared the mutagenicity-emission factors to those of other combustion emissions, some of which are associated with health effects, in order to provide an indication of the relative health impact of the stoves.

Methods

Combustion conditions. Details of the combustion conditions, sources of material, and collection of PM_{2.5} are described by Jetter et al. (2012). In brief, we burned *Quercus rubra* (red oak) with a moisture content of 6% as fuelwood in a TSF, an Envirofit Model G-3300 NDS, and a Philips HD4012 FDS (Figure 1) using a modified water-boiling test protocol (WBT 2013) for all tests that consisted of (a) high power to bring 5 L of water from ambient to boiling temperature and (b) low power to maintain the water temperature at 3°C below boiling temperature for 45 min using a standard 7-L cooking pot as described for all tests (Jetter and Kariher 2009; Jetter et al. 2012). We conducted up to 4 independent burns to assess reproducibility.

The size of the fuelwood was 2 x 2 x 36 cm for the TSF and NDS and 1.5 x 1.5 x 10 cm for the FDS; these sizes were typically used and recommended by the manufacturers. The fuel sizes were matched to the stove, and the comparisons we made were between cookstove systems consisting of the combination of stove, fuel, and operating procedure. Unlike our previous study (Jetter et al. 2012), we tested the NDS without a pot skirt because this configuration is considered more typical of stove use in the field (Adkins et al. 2010). As a reference, we also

combusted propane gas in a Mikachi MNSS 1155 stove under the same conditions as described above.

Collection of emissions and chemical analyses. We collected emissions with a hood and dilution-tunnel system similar to that of Jetter et al. (2012). Briefly, we collected integrated filter samples of PM_{2.5} on 47-mm diameter, 2- μ m thick Teflon[®] filters that were weighed prior to and after collection with a microbalance in an environmentally controlled chamber. Sampling spanned two complete test phases (high-power followed by low-power operation), but we omitted the period between test phases to avoid any emissions released when the fire was extinguished prior to restarting for the second phase.

Emission factors. Emission factors based on useful-energy delivered have denominators with measuring units of megajoule delivered (MJ_d), and those based on fuel-energy used have denominators with units of megajoule thermal (MJ_{th}). Energy efficiency is the ratio of useful-energy delivered to fuel-energy used (MJ_d/MJ_{th}). We calculated pollutant- and mutagenicity-emission factors in a variety of units, and emission factors based on useful cooking energy (MJ_d) enabled comparisons among all cookstove/fuel combinations. For example, we estimated for mutagenicity-emission factors the number of revertants (rev) per MJ_d as:
$$\text{rev/MJ}_d = (\text{rev/mg PM}_{2.5}) \times (\text{mg PM}_{2.5}/\text{MJ}_d).$$

We determined the cooking energy delivered (MJ_d) by the (a) sensible heat that raised the pot water temperature and (b) latent heat that produced steam as described by Jetter et al. (2012). We calculated fuel energy used (MJ_{th}) by the (a) mass of fuel used and (b) fuel-energy content measured with a bomb calorimeter; this value was 18,310 kJ/kg on a dry basis for the lower heating value. We calculated pollutant-emission factors from weighted averages of values measured during high- and low-power operation. These weighted averages were directly

comparable to the emission factors determined from the organic extracts of the PM because we combined filters from both power conditions to have suitable amounts of extract for chemical and mutagenicity analyses. For example, the CO per fuel-energy emission factors were calculated using the equation:

$$CO \text{ per fuel energy } \left(\frac{g}{MJ_{th}} \right) = \frac{CO_{total,HP} + CO_{total,LP}}{E_{th,HP} + E_{th,LP}}$$

Where:

$CO_{total,HP}$	Mass of CO emitted during the high-power phase (g)
$CO_{total,LP}$	Mass of CO emitted during the low-power phase (g)
$E_{th,HP}$	Energy contained in the fuel burned during the high-power phase (MJ)
$E_{th,LP}$	Energy contained in the fuel burned during the low-power phase (MJ)

We measured a variety of pollutants in the emissions in real time, including CO using a non-dispersive infrared analyzer, total hydrocarbons (THC) and methane (CH₄) using flame-ionization detection analyzers, nitrogen oxides (NO_x) by a chemiluminescence analyzer, and BC by an aethalometer. We extracted the organics from the PM_{2.5} with dichloromethane (DCM), determined the percentage of extractable organic material (% EOM) by gravimetric analysis (DeMarini et al. 2004), and determined the concentrations in the extracts of 32 PAHs as described by Kooter et al. (2011) and of levoglucosan as described by Jedynska et al. (2015). We solvent-exchanged the extracts into dimethyl sulfoxide (DMSO) at 2 mg EOM/ml DMSO for the mutagenicity assays.

Mutagenicity assays. We performed the *Salmonella* plate-incorporation mutagenicity assay as described by Maron and Ames (1983) using strains TA98, TA100, TA104, and YG1041 +/- metabolic activation (S9 mix). We used 1 mg of S9 protein/plate with Aroclor-induced

Sprague Dawley rat liver S9 from Moltox (Boone, NC) and incubated the plates for 3 days. Additional details of the procedures and supplies are described in Mutlu et al. (2013). The negative control was DMSO, and the positive controls were 2-nitrofluorene (3 $\mu\text{g}/\text{plate}$) for $-S9$ for strains TA98 and YG1041; sodium azide (3 $\mu\text{g}/\text{plate}$) for TA100 $-S9$; 2-aminoanthracene (0.5 $\mu\text{g}/\text{plate}$) for $+S9$ for strains TA98, TA100, YG1041, and TA104; and methylglyoxal (50 $\mu\text{g}/\text{plate}$) for strain TA104 $-S9$.

We defined a positive mutagenic response as a reproducible, dose-related response with a twofold or greater increase in revertants (rev) per plate relative to the DMSO control. Linear regressions were calculated over the linear portion of the dose-response curves, and the linear portion was defined by the line with the highest r-squared value. We deleted any dose that caused a down-turn in the curve and reduced the r-squared value relative to that produced by inclusion of the lower doses.

Strains TA98 and YG1041 detect mutagens that cause frameshift mutations, and YG1041 has enhanced expression of nitroreductase and acetyltransferase to increase the detection of mutagenic nitroarenes and aromatic amines, respectively. TA100 and TA104 detect mutagens that induce base-substitution mutations, and TA104 detects oxidative mutagens because some of the targets for reversion in this strain are AT base-pairs. With some exceptions due to limited sample, we performed all experiments at one plate/dose in at least two independent experiments.

Calculation of mutagenicity-emission factors. We calculated mutagenicity-emission factors in the same way we calculated the pollution-emission factors described above. The mutagenic potencies (rev/ μg EOM) were first calculated as the slope of the linear portion of the dose-response curves by averaging the data (rev/plate for each dose) from at least 2 independent experiments. These mutagenic potencies (rev/ μg EOM) were then multiplied by the %EOM

(i.e., the EOM fraction, which is the $\mu\text{g EOM}/\mu\text{g PM}$) to give the mutagenic potencies of the mass of particles ($\text{rev}/\mu\text{g particle}$), which is the same as $\text{rev}/\mu\text{g PM}_{2.5}$. These values were multiplied by 1000 to convert them to $\text{rev}/\text{mg PM}_{2.5}$. The resulting mutagenic potencies of the particles ($\text{rev}/\text{mg PM}_{2.5}$) were then converted to units of $\text{rev}/\text{MJ}_{\text{th}}$, $\text{rev}/\text{MJ}_{\text{d}}$, $\text{rev}/\text{kg fuel}$, and rev/h using measured $\text{PM}_{2.5}$ -emission factors reported in this study. These reported values were in units of $\text{mg PM}_{2.5}/\text{MJ}_{\text{th}}$, $\text{mg PM}_{2.5}/\text{MJ}_{\text{d}}$, $\text{mg PM}_{2.5}/\text{kg fuel}$, and $\text{mg PM}_{2.5}/\text{h}$. For example, $\text{rev}/\text{mg particle} \times \text{mg particles}/\text{MJ}_{\text{th}} = \text{rev}/\text{MJ}_{\text{th}}$.

Statistical analyses. We determined mutagenic potencies of the EOM, of the $\text{PM}_{2.5}$, and the mutagenicity-emission factors as follow. We calculated the slope and standard error (SE) from the raw mutagenicity data (rev/plate) to determine the mutagenic potency of the EOM, expressed as $\text{rev}/\mu\text{g EOM} \pm \text{SE}$; we did this using regression models with SAS Proc GLM. We multiplied these mutagenic potencies by 1000 to convert them to $\text{rev}/\text{mg EOM} \pm \text{SE}$, and then we multiplied these by the %EOM, which was a constant, to determine the mutagenic potencies of the $\text{PM}_{2.5}$ ($\text{rev}/\text{mg PM}_{2.5} \pm \text{SE}$). We then converted the $\text{rev}/\text{mg PM}_{2.5}$ to a mutagenicity-emission factor by using the appropriate $\text{PM}_{2.5}$ emission factor ($\text{mg PM}_{2.5}/\text{MJ}_{\text{th}}$, $\text{mg PM}_{2.5}/\text{MJ}_{\text{d}}$, $\text{mg PM}_{2.5}/\text{kg fuel}$, or $\text{mg PM}_{2.5}/\text{h}$) for each stove. These $\text{PM}_{2.5}$ -emission factors were associated with a SE; thus, we calculated the SE for the mutagenicity-emission factors according to Goodman's expression, using the formula $s_{AB}^2 = B^2 s_A^2 + A^2 s_B^2 + s_A^2 s_B^2$. We calculated tests for differences among FDS, NDS, TSF, and the diesel engine mutagenicity-emission factors using Wald statistical tests for each relevant pair. FDS, NDS, TSF, and the diesel engine were considered to be independent; thus, pairwise covariances were assumed to be 0. We calculated Pearson correlation coefficients to compare all the emission factors. We considered results significantly different for $p < 0.05$.

We performed two independent combustion experiments involving mutagenicity measurements for the FDS. Consequently, we performed statistical analyses to compare the variability of 5 pollutant- and 8 mutagenicity-emission factors (all expressed per MJ_d) between these replicate experiments. For this analysis, we reported the experimental parameters and pollutant-emission factors as weighted averages (described in the sub-heading *Emission factors* in Methods) with sample standard deviations (SD), standard errors (SE), coefficients of variation (CV), and number of replicates (n). Among these, carbon dioxide (CO₂) was the primary product of biomass combustion. We used the CO₂ emission factor and the rate at which fuel was consumed (the fuel-burn rate) to compare the reproducibility of the two experiments. We did not use emissions data from Experiment 1 to generate any of the emission factors or mutagenicity data reported elsewhere in the manuscript. Instead, these were included in the statistical analysis only for the purpose of comparing replicate runs of the FDS. Experiment 2 represents the set of replicate experiments using the FDS that we analyzed and report in this study.

We compared the emission factors between the two FDS experiments described above using a two-tailed Student's t-test with Welch's correction. We compared the mutagenicity data using linear regressions for the dose-response curves generated by 2 independent mutagenicity measurements for each of two combustion experiments for a total of 2 experiments. We then compared the slopes of the resulting regressions by using Statgraphics Centurion XVI (Statpoint Technologies, Inc., Warrenton, VA). The Comparison of Regression Lines procedure is designed to compare the regression lines relating Y and X at two or more levels of a categorical factor. Tests were performed to determine whether there were significant differences between the intercepts and the slopes at the different levels of that factor. We plotted the regression lines,

identified unusual residuals, and made predictions using the fitting model. We analyzed the data from the two groups in a multiple-regression model that allowed for a separate intercept and slope for each group. We used a t-test within the model to test for a difference between the two slopes. We considered results significantly different for $p < 0.05$.

Results

Pollutant-emission factors. Weighted averages for high- and low-power test conditions of the pollutant-emission factors determined from continuous-emission monitors as well as the integrated PM_{2.5}, which was collected on filters, are shown in Table 1, and measured parameters from the two individual cooking-power conditions are shown in Table S1. Our results for CO and PM_{2.5} confirmed prior analyses for the NDS and FDS (Jetter et al. 2012). The emissions ranked as follows based on these pollutant-emission factors: Propane < FDS < NDS < TSF; the energy efficiencies ($100 \times \text{MJ}_d/\text{MJ}_{th}$) for these technologies were 67, 36, 32, and 24%, respectively.

Gravimetric analyses of the DCM extracts of the PM collected on the filters showed that the % EOMs were 33.6% for TSF, 17.9% for NDS, 3.0% for FDS, and 0% for propane gas. Because there were no detectable levels of EOM in the propane emissions, we did not subject that extract to chemical analysis. However, analyses of the other extracts resulted in chemical-emission factors for the 16 EPA priority PAHs, the 9 oxy-PAHs, and levoglucosan shown in Table S2. Although we also analyzed these extracts for 6 nitro-PAHs as described (Mutlu et al. 2013), we did not find detectable levels of any of them (data not shown). Using data from Table S2, Figure 2 illustrates that for these PAH-emission factors, the emissions ranked as follows: FDS < NDS < TSF.

Mutagenic potencies of EOM and particles. There was no detectable EOM in the extract of the propane emissions, and the extract was not mutagenic (data not shown); the average rev/plate for the three emissions are shown in Table S3. With only a few exceptions, the mutagenic potencies of the EOM ranked as follows among the strain/S9 combinations: TSF < NDS < FDS (Figure 3A and Table S4 for corresponding numeric data). The highest mutagenic potencies were in strain TA100 +S9, which detects PAHs, and strain YG1041 –S9, which detects nitroarenes.

In contrast to the EOM, the mutagenic potencies of the particles generally ranked in the opposite way as that of the EOM: FDS < NDS < TSF (Figure 3B and Table S5 for corresponding numeric data). However, the mutagenic potencies of the particles in 5 of the 8 strain/S9 combinations were not significantly different between the TSF and NDS (Figure 3B). A sequential decrease in the mutagenic potencies of the particles from all three emissions was found for TA100 +S9 (which detects PAHs), TA104 +S9 (which detects oxidative mutagens), and YG1041 –S9 (which detects nitroarenes). This general reversal of potencies among the stoves between EOM and particles indicates that high-efficiency combustion (FDS) produces organics that are more mutagenic per mass of EOM than does low-efficiency combustion (TSF). However, the particles from high-efficiency combustion (FDS) had only 10% of the amount of EOM compared to particles from low-efficiency combustion, resulting in particles from the FDS that were less mutagenic per mass than those from the TSF.

Mutagenicity-emission factors. Regardless of the units of expression, the strain, or the presence/absence of S9, the emissions ranked as follows based on rev/MJ_d: FDS < NDS < TSF (Table 2); the numerical values are illustrated in Figure 3. The rev/MJ_d were significantly different ($p < 0.001$) among all three emissions within each strain/S9 combination (Figure 3). In

the presence of S9, the largest mutagenicity-emission factors for each emission expressed by any unit were in strain TA100 + S9, indicating an important role for S9-requiring base-substitution mutagens such as PAHs. The second was in YG1041, indicating a role for aromatic amines. The largest values in the absence of S9 were in strain YG1041, indicating an important role for frameshift mutagens such as nitroarenes. Results in strain TA104 with or without S9 indicated a role for oxidative mutagens. The lowest values were in strain TA98 +/- S9, suggesting that frameshift mutagens in addition to or other than nitroarenes also contributed to the mutagenicity of the emissions. For all strains, the rev/MJ_d were reduced by ~50% by the NDS and by >90% by the FDS relative to the TSF (Table 2).

To determine how reproducible the pollutant-emission factors were, we calculated them from a set of multiple combustion experiments with the TSF, NDS, FDS, and propane fuel. Experiments were highly reproducible as indicated by the coefficient of variation (CV) values <10% for most fuel-burn rates and for CO₂ emissions, the primary product of combustion (Tables S6 and S7). Numbers of replicates (n) are shown in the tables. The relatively low CV values (<10%) for the fuel-burn rates and CO₂ emission factors (expressed either as MJ_d or MJ_{th}) indicate that the stoves were operated consistently across all test replicates, resulting in reproducible data. The factors with CV values greater than those of the fuel-burn rates and CO₂ emission factors reflect the inherent variability of the fuel stock and physical processes involved in biomass combustion. With the exception of the emission factors for BC ($p = 0.039$) and mutagenicity in strains TA98 ($p < 0.001$) and YG1041 ($p = 0.013$) of *Salmonella* –S9, there were no significant differences between replicate experiments for the other 6 emission factors for the FDS, including mutagenicity in TA98 and TA100 +S9 (Table S8).

Comparison of emission factors. With the exception of NO_x (Pearson correlations 0.58–0.76), the other emission factors were highly correlated (Pearson correlations ≥ 0.97) (Table S9). Table 3 shows the percent reduction in the emission factors achieved by the two stoves relative to the TSF. Similar results were obtained for emission factors based on continuous-emission monitoring as with those based on extracts of the PM_{2.5}. Excluding NO_x and averaging the percent reduction of the remaining emission factors expressed per MJ_d in Table 3 (and considering that the total PAHs includes the 16 EPA PAHs and the oxy-PAHs), the NDS and FDS reduced these emission factors by 68 and 92%, respectively, relative to the TSF.

Discussion

Pollutant-emission factors. As noted in the Materials section, the comparisons we made were between cookstove systems consisting of stove, appropriate fuel, and operating procedure for each stove. We operated the stove systems as intended by the manufacturers; performance may vary if the systems are not operated as intended. The TSF configuration and thermal efficiency were similar to those in Jetter and Kariher (2009), but the fuel-burning rate was less here because we used a larger sized fuelwood, which is more representative of the average size of fuelwood used in the field (Rosenbaum et al. 2013). The thermal efficiency for the NDS was 38.0% in our previous study and 31.2% here; and the high-power fuel-burning rate for the TSF was 25.4 g/min in our previous study and 15.3 g/min here (Table S1). Our NDS-emission factors for CO and PM_{2.5} were also similar to those in our previous study (Jetter et al. 2012), but the thermal efficiency was less because here we tested the stove without a pot skirt, which is considered to be more typical of operation in the field. The FDS was tested with the same configuration and nearly the same fuel-burning rates as in our previous study (Jetter et al. 2012), and the results were similar.

Emission rates for $PM_{2.5}$ and CO may be used to rank stoves according to tiers of performance for indoor emissions as defined by the ISO (2012) guidelines: Tier 0 represents lowest performance, typical for a TSF, whereas Tier 4 represents an LP/natural gas stove. Results show that many other types of emission factors (THC, CH_4 , BC, PAHs, levoglucosan, and mutagenicity) correlate with $PM_{2.5}$ and CO (Table S9) for the stove and fuel combinations tested under our controlled conditions. Relative to the TSF, the FDS reduced the emission factors associated with products of incomplete combustion by >80% (Table 3); the FDS is rated in Tier 3 for indoor emissions with $PM_{2.5} \leq 480$ mg/h (Jetter et al. 2012).

Other studies have measured as many as 38 or 48 PAHs, along with 3 other pollutant-emission factors (organic carbon, PM, elemental carbon) for various fuels and stoves (Shen et al. 2012, 2013). Here we determined the emission factors for only 32 PAHs, but also for 6 other pollutants, a marker of woodsmoke (levoglucosan), and for the first time mutagenicity for a NDS, FDS, and TSF for comparison. Improved combustion efficiency was associated with reduced emission factors for the various species of PAHs, levoglucosan, and mutagenicity. The high correlations among all of the emission factors other than NO_x were expected because these indicators are all measures of the incomplete combustion of carbon species. In contrast, NO_x is associated with fuel and atmospheric nitrogen and results from complex mechanisms related to flame aerodynamics, fuel-oxidant mixing, and temperature; it is associated only indirectly with carbon burnout during combustion.

Mutagenicity-emission factors. Figure 4A summarizes the published mutagenicity-emission factors (rev/MJ_{th}) in strain TA98 +S9 for a range of combustion emissions, including those from the present study. Data for the cookstoves were expressed as thermal energy (Table 3) rather than as delivered energy in order to permit comparisons because the other emissions are

expressed per unit of fuel energy used. The rev/MJ_{th} were lower for fossil fuels burned in large power-conversion systems employing engineering principles designed to maximize temperatures, fuel and oxidant mixing, flame stability, and combustion efficiency; and higher in smaller, less-efficient systems and open, uncontrolled combustion of poorly characterized fuels. Gaseous, homogeneous fuels produced lower emission factors than solid, heterogeneous fuels—as illustrated also by our data in Tables S5 and S6 for wood versus propane.

For the four emissions for which SE values were known (the diesel generator and the three stoves), all had rev/MJ_{th} values that were significantly different from each other ($p < 0.001$) (Figure 4B). The mutagenicity-emission factor for the most efficient stove (FDS) was significantly less than that of the 4.3-kW diesel generator, but was an order of magnitude greater than that of the 150-kW diesel engine. Thus, the FDS was intermediate to a large diesel engine (from a bus) and a small diesel generator. The rev $\times 10^5$ /MJ_{th} were 0.0276, 0.2, 0.42, 1.23, and 2.42/2.5/2.5 for the diesel engine, the FDS, the diesel generator, the NDS, and the TSF/residential wood fireplace/open burning of agricultural plastic, respectively (Figure 4).

The pollutant-emission factors associated with the propane stove were generally orders of magnitude lower than those associated with biomass burning (Table 1). Likewise, the mutagenicity-emission factor in TA98 +S9 based on fuel- energy used (rev/MJ_{th}) for utility natural gas (20 rev/MJ_{th}) (DeMarini et al. 1992) is 3 orders of magnitude lower than that of the best stove in the present analysis (FDS at 20,000 rev/MJ_{th}) (Table 2). Such results indicate that emissions from the combustion of liquid/gas fuels are considerably less polluting than those from the solid-fuel stoves that we evaluated here.

Consistent with this, a study of pregnant Peruvian women reported that 12 women who cooked with wood plus 4 who cooked with kerosene had higher urinary concentrations of

hydroxylated PAHs (1- and 2- and 3-hydroxy-fluorene; and 2- and 4-hydroxy-phenanthrene) than 27 women who cooked with LP and gas fuels (Adetona et al. 2013a). Such data indicate the importance of substituting wood or kerosene with LP/gas fuels or electricity, improving ventilation, and/or developing cleaner solid-fuel technologies to replace solid-fuel biomass burning for cooking among under-served populations (Balakrishnan et al. 2014; Pachauri et al. 2013).

The greatest reduction in mutagenicity-emission factors was by the FDS relative to the TSF in TA100 +S9 (Figure 3), which detects PAH mutagenicity, consistent with the reductions in PAH-emission factors by the FDS relative to the TSF (Figure 2). Urinary mutagenicity was elevated in 49 Brazilian charcoal workers with high exposure to woodsmoke outdoors compared to 34 workers with no exposure, and this correlated with urinary concentrations of 2-naphthol and 1-pyrenol, of which the parent compounds (naphthalene and pyrene) have been found in woodsmoke (Kato et al. 2004). A study by Long et al. (2014) of Mayan individuals that used traditional wood-fired steam baths reported that urine mutagenicity and exhaled CO both increased after use of the steam bath and were significantly correlated ($r^2 = 0.53$, $p < 0.001$).

Using data for all three emissions (TSF, NDS, FDS), we regressed values of rev of TA100 + S9/MJ_d (55.7, 16.1, and 1.8 rev x 10⁵/MJ_d, respectively) (Table 2) against μg oxy-PAHs/MJ_d (921.9, 282.1, and 101.1 μg/MJ_d, respectively) (Table S2) and estimated a slope of 0.06 x 10⁵ rev/μg oxy-PAHs, which was ~9 times greater than the corresponding estimate for TA100 +S9/MJ_d and the 16 EPA Priority PAHs (0.007 x 10⁵ rev/μg EPA PAHs). The higher mutagenic potency (slope) of the oxy-PAHs further supports the contribution of the oxy-PAHs to the mutagenicity of these emissions.

This is consistent with studies showing that oxy-PAHs are associated with fine PM and cookstove combustion (Shen et al. 2011). In addition, oxidative damage was increased in human lung epithelial and monocytic cell lines exposed *in vitro* to ambient PM from a Danish village with high wood stove use and to PM collected from wood-stove exhaust (Danielsen et al. 2011). Using urinary concentrations of 8-isoprostane (a product of lipid peroxidation) as a biomarker of oxidative stress, Commodore et al. (2013) found an increase in this biomarker in samples of 48-h personal PM_{2.5} exposures among 69 Peruvian women using wood for cooking indoors. Adetona et al. (2013b) found a pre- versus post-shift increase in 8-hydroxy-2'-deoxyguanosine (8-OH-dG) in 3 wildland firefighters with 0-2 years of firefighting experience; 8-OH-dG was also associated with years of firefighting among 17 subjects. Thus, it appears that chronic exposure to woodsmoke elevates oxidative stress biomarkers.

Issues associated with introducing advanced cookstoves. Although the FDS evaluated here has been adopted and used more than another type of FDS (the Oorja) in field studies in India (Mukhopadhyay et al. 2012), systematic reviews have shown that more work is needed for the successful introduction of improved fuels and/or cookstoves (Lewis and Pattanayak 2012; Rehfuess et al. 2014). More than a dozen studies have evaluated indoor-air quality or the health of the residents after interventions involving stoves or chimneys. Although some reported significant improvements in either the indoor-air quality (Fitzgerald et al. 2012; Noonan et al. 2012) or biomarkers (e.g., COHb, exhaled CO, lymphocyte DNA damage, urinary 1-hydroxypyrene, or ST-segment depression) in the residents in the home (Eppler et al. 2013; McCracken et al. 2011; Torres-Dosal et al. 2008) after the intervention, many reported either no improvements or, at best, modest improvements in health outcomes. For example, some showed that although urinary concentrations of 10 hydroxylated metabolites of naphthalene, fluorene,

phenanthrene, and pyrene were reduced, the mean concentrations of some, such as 1-hydroxypyrene, were still in the 95th percentile of the U.S. population based on the National Health and Nutrition Survey 2001-2002 or similar to that of smokers (Li et al. 2011; Riojas-Rodriguez 2011). Installation of chimneys in rural homes in Peru or Guatemala to improve indoor air quality or health outcomes did not result in significant reductions in the frequency of childhood pneumonia (Smith et al. 2011) or in concentrations of CO and PM_{2.5} in the home (Hartinger et al. 2013; Pollard et al. 2014).

Conclusions. To our knowledge, our data are the first to link pollutant- and mutagenicity-emission factors for solid-fuel cookstoves of these types and to compare the mutagenicity-emission factors to those of other combustion emissions. Many emission factors (PAHs, PM_{2.5}, THC, CO, CH₄, BC, levoglucosan, and mutagenicity) correlated highly among each other for the stove and fuel combinations tested under our controlled conditions. Based on fuel-energy used (MJ_{th}), the mutagenicity-emission factor for the most efficient stove (FDS) was intermediate to that of a large diesel bus engine and a small diesel generator (Figure 4). Not surprisingly, that of the TSF was similar to that of other open burning, i.e., residential wood fireplaces and open burning of agricultural plastic (Figure 4). We conclude that without adequate ventilation, even the FDS would result in exposure to highly mutagenic emissions indoors, resulting in poor indoor air quality.

Ultimately, the introduction and reliable supply of electricity and LP/natural gas fuels are essential for making significant improvements in health outcomes for the billions of people who currently have limited or no access to such energy sources for cooking (Pachauri et al. 2013; Smith 2014). While the infrastructure is being developed to supply electricity and LP/natural gas

fuels for this population, introduction of advanced, solid-fuel, vented cookstoves is critical to meeting their intermediate needs.

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Table 1. Pollutant-emission factors expressed as weighted averages for two power levels

Pollutant ^b	Emission factor ^b	Emission source ^a			
		TSF	NDS	FDS	Propane
PM _{2.5}	mg/MJ _d	449.4	198.6	78.3	1.2
	mg/MJ _{th}	106.8	64.0	28.4	0.8
	mg/kg of fuel	1544.9	920.7	454.2	35.2
	mg/h	1396.5	770.2	273.0	4.8
CO	g/MJ _d	11.8	4.4	1.2	0.2
	g/MJ _{th}	2.7	1.3	0.4	0.1
	g/kg of fuel	40.6	20.5	7.2	5.8
	g/h	36.7	17.0	4.2	0.8
THC	g/MJ _d	1.1	0.4	0.1	0.0
	g/MJ _{th}	0.3	0.1	0.0	0.0
	g/kg of fuel	3.9	2.1	0.7	0.6
	g/h	3.5	1.7	0.4	0.1
CH ₄	g/MJ _d	0.3	0.1	0.0	0.0
	g/MJ _{th}	0.1	0.0	0.0	0.0
	g/kg of fuel	1.1	0.4	0.2	0.0
	g/h	0.9	0.3	0.1	0.0
NO _x	g/MJ _d	0.2	0.2	0.1	0.1
	g/MJ _{th}	0.0	0.0	0.0	0.0
	g/kg of fuel	0.6	0.7	0.6	1.4
	g/h	0.6	0.6	0.4	0.2
BC	mg/MJ _d	254.0	68.0	42.3	0.0
	mg/MJ _{th}	62.7	22.0	15.1	0.0
	mg/kg of fuel	868.4	320.2	247.5	0.0
	mg/h	786.2	269.3	146.4	0.0

^aData for PM_{2.5} derived from samples collected on filters; all other data

derived from continuous-emission monitoring. TSF, three-stone fire; NDS, natural-draft stove; FDS, forced-draft stove.

^bPM_{2.5}, particulate material ≤ 2.5 μm in diameter; CO, carbon monoxide; THC, total hydrocarbons; CH₄, methane; NO_x, oxides of nitrogen; BC, black carbon; MJ_d, megajoule energy delivered to the cooking pot.

Table 2. Mutagenicity-emission factors ($\times 10^5$) \pm SE derived from organic extracts of PM^a

Units ^c	Strain	TSF ^b		NDS ^b		FDS ^b	
		+S9	-S9	+S9	-S9	+S9	-S9
Rev/MJ _{th}	TA100	12.7 \pm 0.9	2.2 \pm 0.2	4.6 \pm 0.6	1.4 \pm 0.2	0.7 \pm 0.2	0.2 \pm 0.0
	TA98	2.4 \pm 0.2	1.2 \pm 0.1	1.2 \pm 0.2	0.6 \pm 0.1	0.2 \pm 0.0	0.1 \pm 0.0
	TA104	5.6 \pm 0.4	1.8 \pm 0.2	2.1 \pm 0.5	1.2 \pm 0.2	0.4 \pm 0.1	0.2 \pm 0.1
	YG1041	7.6 \pm 0.8	13.0 \pm 1.0	4.2 \pm 0.7	5.0 \pm 0.7	0.5 \pm 0.1	1.5 \pm 0.4
Rev/MJ _d	TA100	55.7 \pm 4.2	9.6 \pm 0.9	16.1 \pm 5.7	4.9 \pm 1.7	1.8 \pm 0.4	0.3 \pm 0.7
	TA98	10.7 \pm 0.9	5.1 \pm 0.6	4.3 \pm 1.6	2.0 \pm 0.7	0.5 \pm 0.01	0.3 \pm 0.1
	TA104	24.4 \pm 1.7	7.9 \pm 0.1	7.2 \pm 2.9	4.1 \pm 1.5	1.2 \pm 0.3	0.6 \pm 0.2
	YG1041	33.3 \pm 3.5	57.2 \pm 4.5	14.5 \pm 5.3	17.4 \pm 6.2	1.4 \pm 0.4	4.2 \pm 1.0
Rev/kg fuel	TA100	191.4 \pm 16.3	32.9 \pm 3.4	74.4 \pm 9.2	22.6 \pm 2.8	10.6 \pm 2.5	1.7 \pm 0.4
	TA98	36.8 \pm 3.5	17.6 \pm 2.3	20.0 \pm 3.1	9.4 \pm 1.2	3.2 \pm 0.8	2.0 \pm 0.5
	TA104	84.0 \pm 6.7	27.5 \pm 3.7	33.6 \pm 7.4	18.8 \pm 2.5	7.1 \pm 1.8	3.2 \pm 0.9
	YG1041	114.3 \pm 12.7	196.7 \pm 17.4	67.3 \pm 9.7	80.8 \pm 10.4	8.2 \pm 2.3	24.4 \pm 5.7
Rev/h	TA100	173.0 \pm 18.5	29.7 \pm 3.6	62.2 \pm 10.5	18.9 \pm 3.2	6.4 \pm 2.2	1.0 \pm 0.4
	TA98	33.2 \pm 3.8	16.0 \pm 2.3	16.7 \pm 3.2	7.9 \pm 1.3	1.9 \pm 0.7	1.2 \pm 0.4
	TA104	76.0 \pm 7.8	24.8 \pm 3.7	28.1 \pm 7.0	15.7 \pm 2.8	4.2 \pm 1.5	1.9 \pm 0.7
	YG1041	103.3 \pm 13.1	177.8 \pm 19.5	56.3 \pm 10.4	67.6 \pm 11.6	4.9 \pm 1.9	14.6 \pm 5.1

^aThese values were calculated as described according to the formulas in the Methods sub-heading titled *Calculation of mutagenicity-emission factors*. The calculation involved the determination of the slope of the linear portion of the dose-response curve produced by the average of the primary data (rev/plate) from at least 2 independent mutagenicity experiments. These slopes (rev/ μ g EOM) were then multiplied by the %EOM to give the rev/ μ g particle, which were then multiplied by 1000 to give rev/mg particle, which is the same as rev/mg PM_{2.5}. The rev/mg PM_{2.5} values were then multiplied as described in the *Statistics* sub-heading of the Methods by the following PM_{2.5}-emission factors: mg PM_{2.5}/MJ_{th}, mg PM_{2.5}/MJ_d, mg PM_{2.5}/kg fuel, or mg PM_{2.5}/h, which were determined as described in the Methods section sub-heading titled *Emission factors*. The resulting mutagenicity-emission factors are those in this table. All mutagenicity-emission factors were calculated from mutagenic potency data (rev/ μ g EOM) that were positive, i.e., the dose-response reached or exceeded a twofold increase in rev/plate relative to the DMSO control.

^bTSF, three-stone fire; NDS, natural-draft stove; FDS, forced-draft stove.

^cRev, revertant; MJ_{th}, megajoule thermal energy; MJ_d, megajoule energy delivered to the cooking pot.

Table 3. Reduction (%) in emission factors by the two stoves compared with the TSF

Sample collection	Emission factor (/MJ _d) ^b	Average % reduction ± SE ^a	
		NDS ^c	FDS ^c
Integrated-filter sampling continuous-emission monitoring	mg PM _{2.5}	55.8 ± 31.3	82.6 ± 7.1
	g CO	62.7 ± 4.9	89.8 ± 1.1
	g THC	63.6 ± 17.9	90.9 ± 6.5
	g CH ₄	66.7 ± 10.4	100.0 ± 3.5
	mg BC	73.2 ± 7.6	83.3 ± 3.3
	g NO _x	0.0 ^d	50.0 ^d
Extract of PM _{2.5} from filters	μg 16 EPA PAHs	63.9 ^d	95.5 ^d
	μg oxy-PAHs	69.4 ^d	89.0 ^d
	μg Total PAHs	64.4 ^d	94.9 ^d
	Rev x 10 ⁵ in TA100 + S9	72.1 ^d	97.0 ^d
	μg Levoglucosan	87.7 ^d	98.0 ^d
Average ^e		68.2	92.0

^a SE = standard error; the number of samples was generally 4 with the exceptions noted in Table S5.

^bMJ_d, megajoule energy delivered to the cooking pot; PM_{2.5}, particulate material ≤ 2.5 μm in diameter; CO, carbon monoxide; THC, total hydrocarbons; CH₄, methane; BC, black carbon, NO_x, oxides of nitrogen.

^cNDS, natural-draft stove; FDS, forced-draft stove.

^dData insufficient to determine SE.

^eAverage of the percent reduction values omitting those for NO_x, and considering that the Total PAHs includes both the 16 EPA PAHs and the oxy-PAHs.

Figure Legends

Figure 1. From left to right: (a) a 3-stone fire (TSF), (b) an Envirofit Model G-3300 natural-draft stove (NDS) or rocket-type stove, and (c) a Philips HD4012 forced-draft stove (FDS).

Figure 2. Pollutant-emission factors expressed as $\mu\text{g}/\text{MJ}_d$ for the 16 EPA Priority PAHs, the 9 oxy-PAHs, and levoglucosan; data are from Table S2.

Figure 3. Mutagenic potencies of the EOM and particles; data from Tables S4 and S5. Mutagenicity-emission factors; data from Table 2. These values were calculated as described according to the formulas in the Methods sub-heading titled *Calculation of mutagenicity-emission factors*. The calculation involved the determination of the slope of the linear portion of the dose-response curve created by the average of the primary data (rev/plate) from at least 2 independent mutagenicity experiments. These slopes (rev/ μg EOM) were then multiplied by the %EOM to give the rev/ μg particle, which were then multiplied by 1000 to give rev/mg particle, which is the same as rev/mg $\text{PM}_{2.5}$. The rev/mg $\text{PM}_{2.5}$ values were then converted to mutagenicity-emission factors (rev/ MJ_d) by multiplying them by $\text{PM}_{2.5}$ emission factors as described in the *Statistics* sub-heading of the Methods and using the following $\text{PM}_{2.5}$ -emission factors: mg $\text{PM}_{2.5}/\text{MJ}_{th}$, mg $\text{PM}_{2.5}/\text{MJ}_d$, mg $\text{PM}_{2.5}/\text{kg}$ fuel, or mg $\text{PM}_{2.5}/\text{h}$, which were determined as described in the Methods section sub-heading titled *Emission factors*. The resulting mutagenicity-emission factors are in the Table 2. All mutagenicity-emission factors were derived from positive mutagenic potency data (rev/ μg EOM), i.e., the dose-response reached or exceeded a twofold increase in rev/plate relative to the DMSO control. The SE is shown for each histogram, and the horizontal bars represent comparisons between emissions where the p -values were >0.05 and, thus, not significantly different; all other comparisons of the 3 emissions within a strain/S9 condition were significantly different from each other ($p < 0.001$).

Figure 4. (A) Comparison of mutagenicity-emission factors in strain TA98 +S9 (rev/ MJ_{th}) for a variety of combustion emissions. (B) Data for the four emissions for which SE values were available, re-plotted on a linear scale. Data for diesel engines are from Mutlu et al. (2015); data for remaining black bars are from DeMarini et al. (1992, 1994). Data for white bars are from Table 2. We calculated the mutagenicity-emission factor for the 150-kW diesel engine from data

in Turrio-Baldassarri et al. (2004) and first published that value (2.76×10^3 rev/MJ_{th} in TA98 +S9) in Mutlu et al. (2015). We calculated the SE for the 4.3-kW diesel generator using data from Mutlu et al. (2015) by the same method we used here to calculate the SE for the 3 stoves. The rev $\times 10^5$ /MJ_{th} in TA98 +S9 for those values with a calculated SE were 0.42 ± 0.04 for the 4.3-kw diesel generator, 0.20 ± 0.04 for the FDS, 1.23 ± 0.20 for the NDS, and 2.42 ± 0.21 for the TSF. Comparisons among these 4 emissions showed them to be significantly different from each other ($p < 0.001$).



Figure 1.

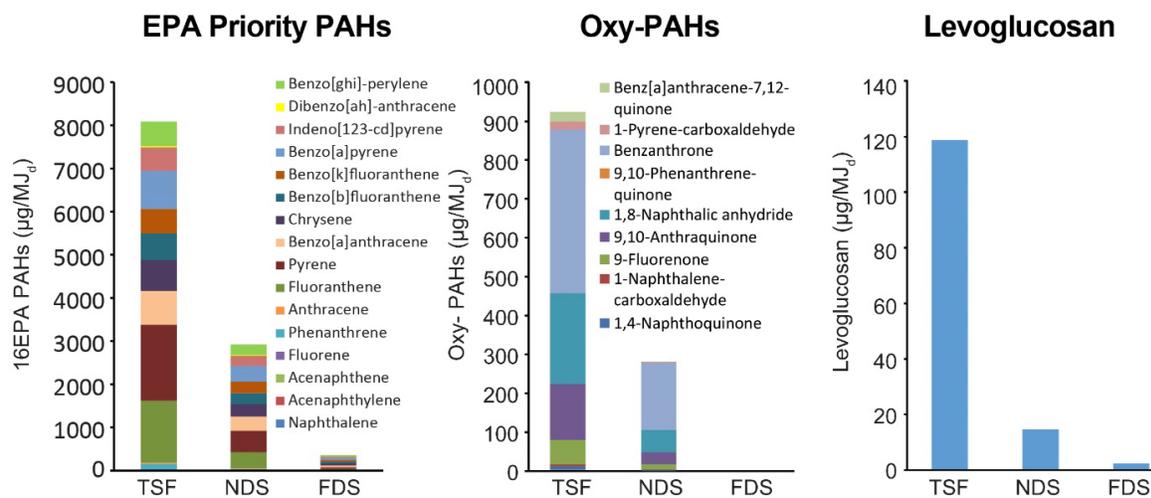


Figure 2.

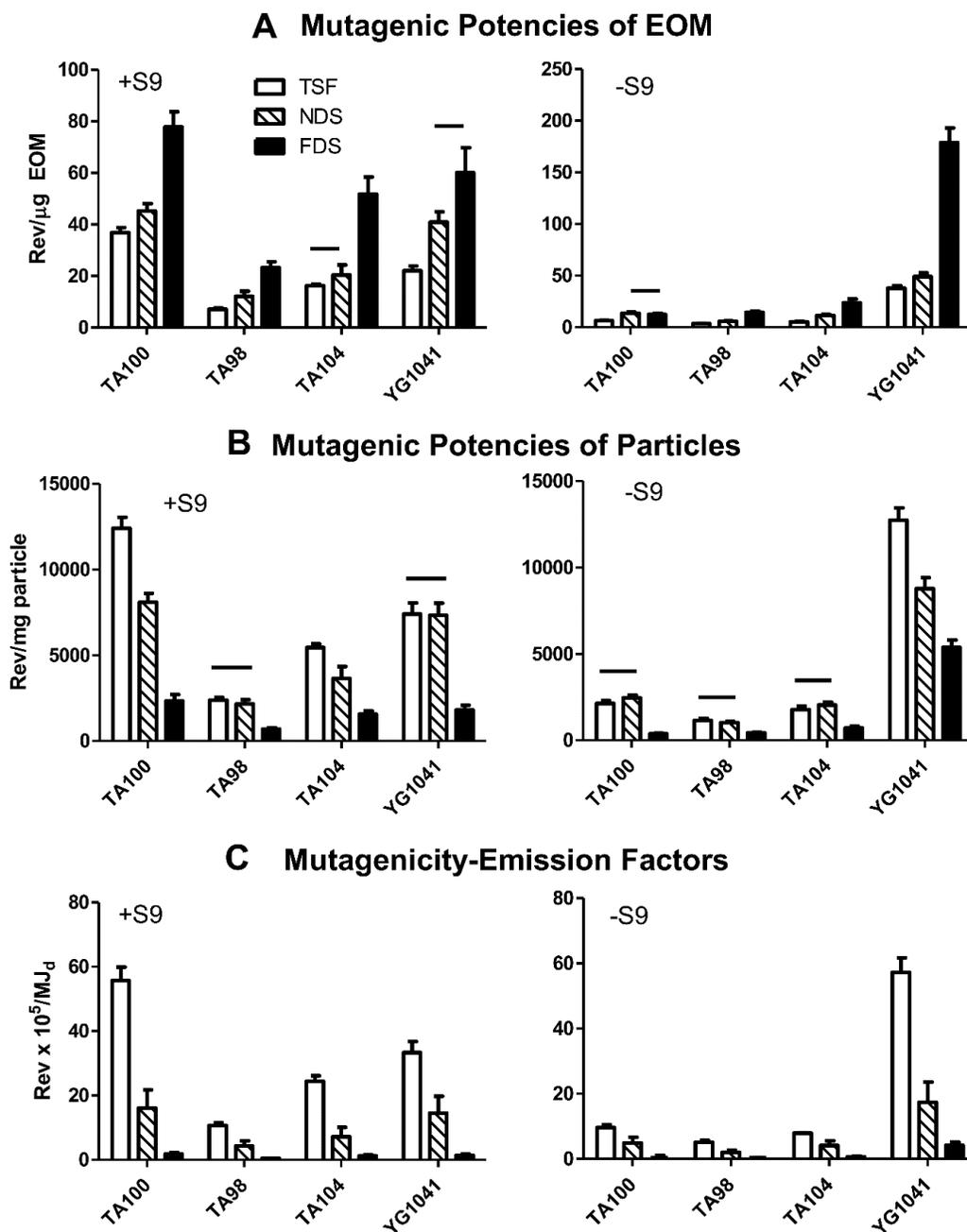


Figure 3.

Figure 4.

