Biodistillate Transportation Fuels 2. – Emissions Impacts

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ABSTRACT

Diesel vehicles are significant sources of NOx and PM emissions, and to a lesser extent, emissions of CO, HC, and toxic species. For many years, biodiesel fuel (and blends of biodiesel) has been promoted as a “clean fuel” alternative to conventional diesel. Based upon previous reviews by EPA, a common understanding has arisen that biodiesel usage reduces CO, HC, and PM emissions significantly, but increases NOx emissions slightly.

This paper discusses a recent review of 94 published reports, from the period of 2000-2008. Assessments were made of the emissions impacts of biodistillate fuels from various engine types, operating conditions, control technologies, and fuel type. In each situation, emissions from the biodistillate case were compared with emissions from a reference diesel fuel case. Graphical displays were developed to show the effects of biodistillate blend level upon 4 emissions species (NOx, CO, HC, PM) from 3 engine types [heavy-duty (HD), light-duty (LD), and single cylinder test engine (TE)].

Results showed that use of biodistillates, even at a 20% blend level, substantially decreased emissions of CO, HC, and PM – generally by 10-20%. Although results varied considerably from one study to the next, similar benefits were seen in both LD and HD engines, regardless of engine technology or test condition. While data were much more limited for renewable diesel cases, these hydropyrolyzed fuels appeared to provide similar emissions reduction benefits for CO, HC, and PM.

NOx emissions impacts were much smaller, and more difficult to discern. Though highly variable, most studies indicated a slight NOx increase when using B100 fuel. For HD engines, the authors’ best estimates are that NOx emissions increase 2-3% with B100, but are unchanged from conventional diesel fuel for B20 blends. Thus, this review indicates smaller NOx effects of biodistillates in HD engines than defined by EPA several years ago. In LD engines, NOx effects appear to be somewhat larger, with increases of 10-15% observed when using B20 and B100, respectively. More sophisticated statistical analyses are required to assess the significance of these small effects.

INTRODUCTION AND BACKGROUND

This effort is part of a larger study sponsored by the Coordinating Research Council (CRC), with the overall objective of assessing the state of knowledge regarding biofuels as blending materials for ultra-low sulfur diesel (ULSD) fuel in transportation applications. Besides emissions impacts, the entire study dealt with policy drivers, feedstocks, fuel production technologies, fuel properties and specifications, in-use handling and performance, and life-cycle impacts. Companion papers address these non-emissions topics.

In this paper, the comprehensive term, biodistillate, is used to include all plant- and animal-derived middle distillate fuels intended for use in diesel engines, regardless of the production technology used to manufacture the fuels. The two major biodistillate categories are:
1. Biodiesel: Fatty acid methyl esters (FAME) produced by transesterification of triglycerides (from animal fats and vegetable oils) with methanol.

2. Renewable diesel: Hydrocarbon fuel produced by catalytic hydrotreatment of the same triglyceride feedstocks.

Other fuel terms such as “1st Generation” and “2nd Generation” are commonly used, but have variable meanings. In this paper, conventional biodiesel (FAME) is regarded as 1st Generation, while renewable diesel is regarded as 2nd Generation.

An earlier review of the impact of B20 on dynamometer-based emissions conducted by EPA[4] reported substantial decreases in HC, CO, and PM emissions, and slight increases in NOx emissions (see Table I and Figure 1). These findings raised serious questions as to the potential impact on NOx emissions following the introduction of biodistillate fuels. More recently, McCormick et al.[5] performed an analysis using updated information and concluded that, on average, there was no net increase in NOx emissions when using B20. (Table I). Both studies reported significant reductions in CO, HC, and PM emissions.

Table I. Average percent change in emissions from use of B20 in HD dynamometer tests

<table>
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<tr>
<th></th>
<th></th>
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</tr>
</thead>
<tbody>
<tr>
<td>NOx</td>
<td>+2.0</td>
<td>+0.6*</td>
</tr>
<tr>
<td>CO</td>
<td>-11.0</td>
<td>-17.1</td>
</tr>
<tr>
<td>HC</td>
<td>-21.1</td>
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</tr>
<tr>
<td>PM</td>
<td>-10.1</td>
<td>-16.4**</td>
</tr>
</tbody>
</table>

*Reported as statistically insignificant  
**Excludes engines equipped with DPF.

Figure 1. Average emissions impacts of biodiesel used in heavy-duty (HD) highway engines (EPA, 2002).

This discrepancy in NOx emissions results between these previous studies is a critical issue that needs to be resolved, since any potential increase in NOx could limit the use of biodistillate fuels. Using the extensive literature review compiled as part of the broader study, this paper investigates the range of reported biodistillate emissions results to develop a better understanding of the true impacts of these fuels.

**ANALYSIS METHODOLOGIES**

Approximately 1000 literature papers and reports compiled as part of the overall CRC study were reviewed for emissions data. The references containing emissions data are given in Appendix V of Hoekman et al.[1] which also provides summary information about each study. This literature review focused on the years 2000 to 2008. Most relevant emissions data have been published within the past five years.

A total of 94 references were identified, which reported 346 distinct emissions test results for different engine types [heavy-duty (HD), light-duty (LD), and single-cylinder test engines (TE)], blend levels (B20, B50, B100, etc.), biodistillate sources (soy oil, rapeseed oil, etc.), and test conditions (steady-state and transient). The few cases using medium duty (MD) engines were lumped with the HD cases.

Absolute emissions rates vary greatly from one test to another, due to large influences of engine type, test cycle, control technology, and other factors. Therefore, the percentage change in emissions when using a biodistillate vs. a reference diesel fuel was the metric chosen for analysis, rather than absolute emission rates. This approach more clearly identified the impacts of specific biodistillate blends on emissions. Publications that did not include a reference diesel fuel for comparison were not used in this evaluation. Also, extreme outliers were omitted from the analysis based on meeting either of the following two simple criteria:

1. In studies reporting a single biodistillate and reference fuel result, did the biodistillate emissions case exceed the reference case by more than 250%?

2. In studies reporting several biodistillate and reference fuel results, did the average of the biodistillate cases exceed the reference case by more than 150%?

Several other limitations of the analysis methodology should be noted. For example, biodistillate and reference diesel fuels varied from one study to the next. ASTM has defined sets of technical standards with which all diesel fuel in the U.S. must comply. However, these standards consist of a series of tests that define minimums, maximums and ranges of properties, with very few exact values. Because of this, compositional variations in fuels are possible, even though all fuels meet the same ASTM specifications.
In addition, different fuel specifications are applied in different countries. In the U.S., conventional diesel fuel is defined by ASTM D 975, while in Europe it is defined by EN 590. Biodiesel (B100) is defined by ASTM D 6751 in the U.S., and by EN 14214 in Europe. ASTM has recently established D 7467 to define B6-B20 blends of biodiesel. These standards have evolved over the years as scientific research and in-use experience have shown the need for greater control of certain properties to ensure satisfactory fuel quality and performance. In this study, published emissions results from all countries and all biodistillate blends were accepted, regardless of the fuel specifications in place at the time.

Significant advancements in engine and emissions control technology have occurred over time. However, there is not a direct correlation between model year and publication date. Many studies did not report the model year as a characteristic of the test engine. (In some cases, old technology systems may be used, even though the publication is very recent.) This lack of information about technology of the test engines also introduced variability into the analysis.

Typically, engine output is controlled with a dynamometer that is either placed on the vehicle's wheels or connected directly to the engine. Due to mechanical losses, the same load applied to the wheels will result in a higher output directly from the engine. This effect could also cause differences in the emissions results. In this study, results from both engine dynamometer and chassis dynamometer tests were included, although they were analyzed separately.

Additionally, several engine test cycles are commonly employed for emissions testing. While there are standard test cycles, such as CFR Title 40 part 86 (On-Highway Heavy-Duty) and JC08 (designed for city traffic), most published emissions results were determined under steady state operation, with constant speeds and loads. Variations in these test conditions can significantly affect an engine's output.

Finally, the literature contains emissions results from a wide variety of biodistillate types. In this review, no attempt was made to distinguish emissions results on the basis of biodistillate feedstock (e.g. soy, rapeseed, algae, etc.). Reported results from all biodistillates were included, even fuels that were prepared from rather exotic, experimental feedstocks.

Reported blend levels ranged from B5 to B100 – though the most commonly studied levels are B20 and B100. Table II shows the number of data points used in assessing the emissions impacts of biodistillate fuels in each engine category.

### Table II. Number of data points used in analysis of biodistillate emissions effects

<table>
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<tr>
<th>Engine Class</th>
<th>Pollutant</th>
<th>B20</th>
<th>B100</th>
<th>Other Blends</th>
<th>Outlier Points</th>
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</table>

### RESULTS AND DISCUSSION

The process of sorting the data and eliminating outliers left a data set of the percentage change in emissions from a biodistillate fuel test compared to a reference diesel fuel test for 4 pollutants (CO, HC, PM and NOx) and 3 classes of engines, (HD, LD and TE.) [PM results included only gravimetric-based measurements (TPM or PM10), not opacity or smoke measurements.] These data were analyzed and displayed in two basic ways to explore: (1) the influence of biodistillate blend level upon criteria emissions and (2) the influence of engine model year upon criteria emissions.

### INFLUENCE OF BIODISTILLATE BLEND LEVEL

To display the influence of biodistillate blend level upon emissions, the data were plotted in a series of three graphs for each engine type, as follows:

1. Data points are shown for averages of each reported test in a given study at a given biodistillate level. This assessment is designed to illustrate variability in the reported emissions test results.
2. Data points are shown for averages of all tests from all studies at a given biodistillate level, with error bars representing the minimum and maximum percent change. This simplifies the previous representation by collapsing several test results to a single point for each blend level.
3. Using the average dataset from above, a best-fit logarithmic trend line was developed for each emissions species as a function of biodistillate level. This representation is similar to what was used in the 2002 EPA document.

The series of graphs developed from this approach are displayed in Figures 2, 3, and 4 for HD, LD, and TE cases, respectively. The top panel in each figure shows a single data point for the average test result at a given biodistillate level, and is color coded by pollutant (NOx, CO, PM and HC). To display the full range of observations, the y-axis spans a percent change of +/-100%. A linear trend line for each of the species is
included, which provides an assessment of the overall change in emissions with increasing levels of biodistillate.

The middle panel collapses the data from the top panel by displaying the average of all test results at a given blend level vs. blend level. Error bars represent the minimum and maximum percent change from a reference diesel fuel for all test results at a specific biodistillate blend level. (These data points and error bars are offset slightly along the x-axis to provide better graphical clarity.) Again, they are color coded based on the four pollutants, and a linear trend line is displayed for each of the species.

The bottom panel in each figure uses the same dataset as the middle panel, but displays a logarithmic trend line based on the average of all emissions for a given biodistillate level. (Note that for this case, the range in the y-axis is from +20% to -40%) This is the simplest way of showing the trend in emissions for each pollutant as a function of biodistillate level, and allows for better comparison with the trends reported in previous reviews.

For all three approaches, trend lines were extrapolated beyond the data set to a B5 level, but were not force fit through zero (i.e., 0 % change for 0 % biodistillate). The location of the trend line at the lowest biodistillate levels should be treated with caution. This is especially true for the logarithmic results, where both the magnitude and direction of the trend line are primarily determined by the lowest % biodistillate data points. A thorough statistical analysis of these data was beyond the scope of our study.

Using the information presented in Figures 2-4, a number of observations regarding the potential impacts of biodistillate blend levels on emissions can be made, with the caveat that this approach does not allow for the assessment of the impacts of specific biodistillate sources, test cycles, control technologies, operating conditions, or model years. A further limitation of this approach is that by not evaluating specific biodistillate sources, a linkage between biodistillate fuel properties and emissions cannot be determined. Given these limitations, we make the following observations regarding the trends in emissions effects across the range of engines, fuels, and test cycles used:

Heavy-Duty Engines (including medium-duty):
- NOx emissions differ very little from reference diesel fuel. The effects at B20 and below are indistinguishable from zero. A slight NOx increase (2-3%) may occur with B100.
- CO, HC, and PM are decreased for all B levels, and decrease further with increasing % B.
- There is a greater impact of % B on HC and PM than on NOx and CO.

Light-Duty Engines:
- NOx emissions are elevated for all B levels, and increase slightly with increasing % B.
- The magnitude of the NOx increase is greater than for the HD case.
- CO, HC, and PM decrease for all B levels, but there is less of an impact of % B on emissions than for HD engines.
- There is a greater impact of %B on HC and PM than on CO.

Single Cylinder Test Engines:
- All emission species decrease with use of biodistillate at all blend levels.
- NOx emissions increase slightly with increasing % B.
- CO and HC emissions are relatively flat and do not appear to be greatly influenced by % B.
- PM emissions decrease with increasing % B.

The overall appearance of these trend lines is similar across all three engine types, exhibiting elevated NOx emissions and reduced CO, HC, and PM emissions with increasing biodistillate content. However, the magnitude of the effects varied somewhat from one engine type to the next. In most cases, the PM effect appeared to be strongest, while the NOx effect was weakest. The range of effects among the four pollutant species was narrower with the TE cases. This may be due to greater control over engine operating parameters, and greater reliance on steady-state conditions (i.e., fewer transients).

To compare the results of these analyses with the findings of other reported studies, the regression equations derived from the logarithmic trend lines were used to predict the % change in emissions of a given pollutant for a specified % B. These regression equations were determined for two different data sets:

1. Full data set: included results from both biodiesel and renewable diesel fuels.
2. Partial data set: included only results from biodiesel fuels. This data set was slightly smaller, with 0-3 data points eliminated, depending upon the engine type and pollutant species investigated.

The emissions impacts of biodistillates at the 20% and 100% blend levels are shown in Table III. Results from the two different data sets are similar. This is to be expected, since very few data points were eliminated in the “biodiesel only” data set. (In fact, the two data sets are identical for the single cylinder test engines.) With HD engines, the “biodiesel only” data set gave slightly larger emissions reduction benefits for CO, HC, and PM. However, the differences are small, and more sophisticated statistical analyses would be required to determine whether they are significant.
Figure 2. Effects of Biodistillate Blends on Exhaust Emissions from HD Engines
Figure 3. Effects of Biodistillate Blends on Exhaust Emissions from LD Engines
Figure 4. Effects of Biodistillate Blends on Exhaust Emissions from Test Engines
Table III. Predicted percent changes in emissions using B20 and B100

<table>
<thead>
<tr>
<th>Pollutant</th>
<th>Full Data Set</th>
<th>Biodiesel Only Data Set</th>
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<tbody>
<tr>
<td></td>
<td>HD</td>
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<tr>
<td>B20</td>
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<td>NOx</td>
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<td>-16.6</td>
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<td>B100</td>
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<td>HC</td>
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<tr>
<td>PM</td>
<td>-36.8</td>
<td>-31.7</td>
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</tbody>
</table>

Notes: HD = heavy-duty and medium-duty CI engines
LD = light-duty CI engines
TE = single cylinder CI test engines

Going from B20 to B100 reduced emissions of CO, HC, and PM for all three engine types. NOx results are less clear. This assessment of HD NOx results suggests that biodistillates have no effect at low levels (B20), but increase NOx slightly (2-3%) at B100 levels. LD results suggest a more consistent NOx increase of 10-15% for B20 and B100, respectively, though the high variability in the emissions results makes these conclusions questionable. For the single cylinder TE cases, NOx emissions appear to be reduced slightly at a B20 level, but not at a B100 level. More sophisticated statistical analyses are required to assess the significance of these small effects.

The HD results for all blend levels reported in this study can also be compared with the EPA (2002) findings that were shown in Fig. 1. Overlaying the data from Fig. 2 (bottom panel) with those of Fig. 1 yields the chart shown in Fig. 5. The solid lines represent findings from this work, while the dashed lines represent the EPA (2002) findings. It is important to remember these data represent a wide collection of engines and control technologies, model years, biodistillate sources, and test cycles, with the EPA (2002) observations reflecting an older set of experiments and engines. Given this caveat, the findings from the two data sets are quite similar, with NOx emissions increasing and CO, HC, and PM emissions decreasing as the biodistillate level increases.

Figure 5 also shows that the percent changes in emissions for all pollutants are lower from this study than in the previous report, i.e. the overall fuel effects are smaller in this study compared to the EPA report. One possible explanation is that emissions from newer engines (and more advanced control technologies) included as part of the current study are lower overall, leading to a reduced impact from the introduction of biodistillate blends.

As a final point of reference, the B20 HD results from this study were compared with those determined by EPA (2000) and McCormick et al. (2006). The comparisons are shown in Table IV. Overall, the predictions for all species are quite similar. Based upon these findings, it is concluded that use of biodistillate blends at a 20% level has a positive impact on diesel CO, HC, and PM emissions, with little impact on NOx emissions. Perhaps the most valid conclusion regarding NOx is that offered in a recent NREL report, “…examination of the NOx results shows that the effects of biodiesel can vary with engine design, calibration, and test cycle. At this time, the data are insufficient for users to conclude anything about the average effects of B20 on NOx, other than it is likely very close to zero.”

Table IV. Comparison of average percent change in emissions from HD dynamometer tests using B20

<table>
<thead>
<tr>
<th>Pollutant</th>
<th>EPA, 2002</th>
<th>McCormick et al., 2006</th>
<th>This Study</th>
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<tbody>
<tr>
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<td>Biodiesel Only</td>
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<td>+0.6*</td>
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<td>PM</td>
<td>-10.1</td>
<td>-16.4**</td>
<td>-15.5</td>
</tr>
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</table>

*Reported as statistically insignificant.
** Excludes engines equipped with DPF.

INFLUENCE OF MODEL YEAR

The effect of engine model year – and hence engine and emissions control technology – upon emissions impacts from biodistillates is of particular interest. Newer engines, with improved emissions control systems and lower overall exhaust emissions could affect the observed impacts of biodistillates. However, as mentioned previously, most publications reporting emissions results from use of biodistillate fuels did not indicate the model year of the test engine. Also, in some cases, the engine(s) employed had been modified in various ways so that direct connection to a particular model year was no longer possible. To address this situation, publication date was used as a rough surrogate for engine model year. Given the limitations mentioned above, it is clear that publication year is a poor surrogate for model year. Nevertheless, this approach provides clearer observation of engine-to-engine and study-to-study variability of the fuel effects at a given blend level.
Two graphs were created for each of four pollutants (NO\textsubscript{x}, CO, HC, and PM); one for B20, the other for B100. Each emissions test was represented by a data point for the average change in emissions reported in the published study. Error bars represent the minimum and maximum reported values. The data were further sorted by engine size (HD, LD or TE), NO\textsubscript{x} emissions control system (yes/no = dotted/solid error bars), and testing procedure (engine or chassis dynamometer test). This approach provides ready observation of variability in the reported data, with the caveat that there is no differentiation among biodistillate type (i.e. soybean, rapeseed, algae, etc.), test cycles, or operating conditions.

The graphs of % change in emissions at fixed biodistillate levels (B20 and B100) for a given publication year are shown in Figs. 6-9. (For improved graphical clarity, the data points and error bars are offset slightly along the x-axis.) Given the limited results for each year, coupled with large error bars associated with most data points, it is difficult to draw quantitative conclusions. However, qualitatively it appears that the % change in all emissions (for both B20 and B100) is largely unaffected across the range of model years and technologies included in these data.

Also shown in Figures 6-9 are separate data points, (indicated by open symbols) for renewable diesel fuel cases. At the present time, there are very few published reports of emissions effects of renewable diesel fuels – only 4 points are included in the B20 plots, and a single point in the B100 plots. Based upon such scant information, it is not possible to draw definitive conclusions. Although PAH, nitro-PAH, and other toxics are of interest, the literature does not contain sufficient emissions data to draw meaningful conclusions about these species. A total of seven recent papers contained information on changes in aldehyde emissions compared with a reference diesel fuel. While some of the papers contained results for speciated aldehydes, others only reported total aldehydes.

NO\textsubscript{x} Emissions (Fig. 6)
- Most HD NO\textsubscript{x} emissions results (for both B20 and B100) lie very close to (or overlap) the zero line.
- Most LD NO\textsubscript{x} emissions results show an increase with both B20 and B100.

CO Emissions (Fig 7)
- The majority of all CO results show clear reductions with use of both B20 and B100.
- B20 and B100 provide CO reduction benefits of approximately equal magnitude.

HC Emissions (Fig. 8)
- The majority of all HC results show clear reductions with use of both B20 and B100.

PM Emissions (Fig. 9)
- Very little PM data are available from single-cylinder test engines.
- The majority of all PM results show clear reductions with use of both B20 and B100.
- B100 generally gives larger emissions reduction benefits compared to B20.

INFLUENCE ON TOXICS EMISSIONS

To address the impact of biodistillates upon emissions of toxics, the database was reviewed for emissions data beyond the criteria species of NO\textsubscript{x}, CO, HC, and PM. The findings were limited, with most observations being for aldehydes. A total of seven recent papers contained information on changes in aldehyde emissions compared with a reference diesel fuel. While some of the papers contained results for speciated aldehydes, others only reported total aldehydes.

The results for formaldehyde, acetaldehyde and total aldehydes are presented graphically in Figure 10. The previously described methodology to evaluate criteria pollutant emissions was also applied in this case. Due to the limited number of data points, Figure 10 includes results from both HD and LD engines, along with a logarithmic fit to the complete data set. Overall, these results suggest a decrease in emissions with increasing blend level; although a few studies reported increasing emissions for B20. This effect is somewhat surprising since biodiesel, which consists of oxygenated species (FAME), might be expected to increase aldehyde emissions.
Figure 6. NOx Emissions for Biodistillate Fuels Compared to Reference Diesel Fuel
Figure 7. CO Emissions for Biodistillate Fuels Compared to Reference Diesel Fuel
Figure 8. HC Emissions for Biodistillate Fuels Compared to Reference Diesel Fuel
Figure 9. PM Emissions for Biodistillate Fuels Compared to Reference Diesel Fuel
CONCLUSIONS

Reduction of exhaust emissions has been one of the drivers for biodiesel fuels for many years. Compared to conventional petroleum-derived diesel fuel, most literature reports indicate 10-20% reductions in CO, HC, and PM emissions when using B20 blends, with larger benefits at higher blend levels. Similar benefits were observed with both LD and HD engines/vehicles, though individual results varied widely from one study to the next. Although data are much more limited for renewable diesel cases, it appears that similar benefits in reduction of CO, HC, and PM were observed with these hydroprocessed fuels.

The fuel effects upon NOx emissions were much smaller, and difficult to discern. This review of HD NOx results suggests that biodistillates have no effect at low levels (B20) but increase NOx slightly (2-3%) at B100 levels. LD results suggest a more consistent NOx increase of 10-15% for B20 and B100, respectively, though the high variability in these emissions results makes such conclusions somewhat questionable. Also, due to the high variability, it is not possible to discern clear trends in fuel effects with changes in engine technology or model year. Based upon the small amount of available information, it appears that the NOx emissions effects of renewable diesel are not greatly different from the effects of biodiesel. More work is needed in this area to assess whether significant differences exist.

Much less information is available regarding non-criteria pollutant emissions from use of biodistillate fuels. Most mobile source air toxics (MSAT) data relate to formaldehyde and acetaldehyde. As with the criteria pollutants, these aldehyde results are highly variable, showing both increases and decreases compared to use of conventional diesel. Many (but not all) results from recent studies indicate a slight decrease in aldehyde emissions when using B20, and a larger decrease when using B100. A more substantial body of emissions data is necessary to confirm (or refute) these observations using modern engines and fuels.

ACKNOWLEDGMENTS

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REFERENCES


