

1     **AIR QUALITY IMPACTS OF INCREASED USE OF ETHANOL UNDER THE**  
2             **UNITED STATES' ENERGY INDEPENDENCE AND SECURITY ACT**

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23

1 **Abstract**

2

3           Increased use of ethanol in the United States fuel supply will impact emissions  
4 and ambient concentrations of greenhouse gases, “criteria” pollutants for which the U. S.  
5 EPA sets ambient air quality standards, and a variety of air toxic compounds. This paper  
6 focuses on impacts of increased ethanol use on ozone and air toxics under a potential  
7 implementation scenario resulting from mandates in the U. S. Energy Independence and  
8 Security Act (EISA) of 2007. The assessment of impacts was done for calendar year  
9 2022, when 36 billion gallons of renewable fuels must be used. Impacts were assessed  
10 relative to a baseline which assumed ethanol volumes mandated by the first renewable  
11 fuels standard promulgated by U. S. EPA in early 2007. This assessment addresses both  
12 impacts of increased ethanol use on vehicle and other engine emissions, referred to as  
13 “downstream” emissions, and “upstream” impacts, i.e., those connected with fuel  
14 production and distribution. Air quality modeling was performed for the continental  
15 United States using the Community Multi-scale Air Quality Model (CMAQ), version 4.7.  
16 Pollutants included in the assessment were ozone, acetaldehyde, ethanol, formaldehyde,  
17 acrolein, benzene, and 1,3-butadiene. Results suggest that increased ethanol use due to  
18 EISA in 2022 will adversely increase ozone concentrations over much of the U.S., by as  
19 much as 1 ppb. However, EISA is projected to improve ozone air quality in a few  
20 highly-populated areas that currently have poor air quality. Most of the ozone  
21 improvements are due to increases in nitrogen oxides (NO<sub>x</sub>) in volatile organic compound  
22 (VOC)-limited areas. While there are some localized impacts, the EISA renewable fuel  
23 standards have relatively little impact on national average ambient concentrations of most

1 air toxics, although ethanol concentrations increase substantially. Significant  
2 uncertainties are associated with all results, due to limitations in available data. These  
3 uncertainties are discussed in detail.

4

## 5 **1. Introduction**

6

7 In recent years, use of ethanol as a component of vehicle fuel has been heavily  
8 promoted for a variety of reasons, including reduction of reliance on fossil fuel, reduction  
9 in emissions that lead to global warming, and reduction in ambient concentrations of a  
10 variety of air pollutants. Changes to the U. S. EPA’s Renewable Fuel Standard program  
11 (RFS2), as mandated by the U. S. Energy Independence and Security Act of 2007, will  
12 result in large increases in the amount of ethanol used. Whether increased use of ethanol  
13 made from various feedstocks and production pathways results in net increases or  
14 decreases in greenhouse gases is currently the subject of considerable scientific debate  
15 (Niven, 2005; Crutzen et al., 2008; Wakely et al., 2009). In addition, increased use of  
16 ethanol will impact “criteria” pollutants (e.g., ozone) for which the U. S. Environmental  
17 Protection Agency (U. S. EPA) has set National Ambient Air Quality Standards  
18 (NAAQS), as well as those pollutants referred to as “air toxics” based on their potential  
19 to cause adverse cancer and non-cancer health effects. This paper focuses on these two  
20 sets of pollutants and how increased use of ethanol could potentially impact their  
21 emissions and concentrations in the ambient air. Impacts of ethanol in fuel on vehicle  
22 and nonroad engine emissions are addressed, as well as impacts associated with fuel  
23 production and distribution. Concerns that increased use of ethanol could lead to higher

1 volatile organic compound (VOC) emissions and elevated tropospheric ozone levels will  
2 be addressed in this paper (Jacobson, 2007; Smith et al., 2009).

3  
4       Limitations in data present challenges in assessing impacts of increased ethanol  
5 use. Most available engine emissions data for ethanol blends are from gasoline highway  
6 vehicles using ten percent ethanol by volume (E10) blends with limited data on 85  
7 percent ethanol (E85) blends. It has long been known that addition of 10 percent ethanol  
8 by volume increases vehicle emissions of acetaldehyde substantially (U. S. EPA, 1993).  
9 Most studies show reductions in emissions of carbon monoxide (CO), benzene, and 1,3-  
10 butadiene (U. S. EPA, 1993; U. S. EPA, 2007a), but there are other studies that do not  
11 (Durbin et al., 2007). Inconsistencies across studies can result from differences in vehicle  
12 emission control technologies, fuel composition, test cycle, and age of vehicles. Impacts  
13 on emissions of VOCs and oxides of nitrogen (NO<sub>x</sub>) are clearly dependent on vehicle  
14 control technology (U. S. EPA, 2007a). Compared to gasoline without ethanol (E0), data  
15 from recent E85 testing consistently show reductions in emissions of benzene and 1,3-  
16 butadiene, increases in formaldehyde, and very large increases in acetaldehyde (Yanowitz,  
17 et al., 2009; Graham et al., 2008; Southwest Research Institute, 2007). Generally, data  
18 also show CO reductions but are more equivocal for NO<sub>x</sub> and VOC. When evaluating  
19 impacts of ethanol on vehicle emissions, it is important to characterize direct emissions of  
20 ethanol, which have the potential to cause adverse health effects and can also react in the  
21 atmosphere to form acetaldehyde and peroxyacetyl nitrate (PAN). Other emissions  
22 resulting from increased levels of ethanol in the fuel can also react in the atmosphere to  
23 form ozone, organic aerosols, acetaldehyde and other pollutants. Accurate

1 characterization of emissions of these pollutants is critical in characterizing air quality  
2 impacts of increased ethanol use.

3

4       Along with the impacts of increased ethanol use on vehicle and other engine  
5 emissions, there are “upstream” impacts, i.e., those connected with fuel production and  
6 distribution. These impacts come from changes in agricultural processes, feedstock  
7 transportation, and the production and distribution of renewable fuel. These processes  
8 occur domestically and internationally. Emissions related to agricultural processes can  
9 come from farm equipment, fertilizer production and application, pesticide production  
10 and application, burning of crop residue, and fugitive dust from field tilling and related  
11 activities. Feedstocks may be transported by rail, marine vessels, or trucks, and the  
12 emissions associated with this transport must be estimated. In addition, there are  
13 significant emissions associated with ethanol production facilities, as well as from the  
14 production of energy used in the operation of these facilities. Key pollutants emitted by  
15 these facilities and reported in U. S. EPA’s Toxics Release Inventory and National  
16 Emissions Inventory (NEI) include CO, particulate matter (PM), NO<sub>x</sub>, sulfur oxides  
17 (SO<sub>x</sub>), and VOCs. VOCs emitted in the largest quantity from ethanol plants include  
18 ethanol and acetaldehyde (Brady and Pratt, 2007). However, displacement of gasoline  
19 with ethanol can result in reduced emissions at petroleum refineries. Recent research  
20 suggests that emissions associated with the transport and distribution of ethanol and  
21 gasoline/ethanol blends may be significant as well (Wakeley et al., 2009). These  
22 emissions result from combustion of fuels used in transport and distribution, as well as  
23 evaporative and spillage emissions from storage and transfer activities. Since a pipeline

1 system does not exist for ethanol, increased ethanol use is likely to increase emissions  
2 associated with distribution of fuels.

3

4         Analyses done to date on impacts of increased ethanol use either model the  
5 impacts from extreme cases or do not account for impacts from all sources across the life-  
6 cycle, or both. For instance, one recent analysis modeled emissions impacts, air quality  
7 and potential health risks from changes in downstream sources due to 100 percent  
8 penetration of E85 (Jacobson, 2007). While this study accounted for only impacts of  
9 changes in vehicle and nonroad equipment emissions, subsequent work (Jacobson, 2009)  
10 suggested downstream sources had a larger impact on mortality than upstream sources.  
11 However, it should be noted that Jacobson used upstream emissions from Delucchi  
12 (2006), which does not account for all upstream emission sources that can be impacted by  
13 the increased use of ethanol. Also, while 100 percent penetration of E85 is not realistic,  
14 such an extreme case can be used to scale impacts for different E85 penetration levels.

15

16         Qualitative inferences of the likely impacts of increased ethanol use can be  
17 obtained from analysis of existing air quality monitoring data in locations where ethanol  
18 use has increased. Much early work focused on air quality impacts of increased use of  
19 ethanol from sugar cane in Brazil. Brazil is the only country in the world where a  
20 nationwide, large-scale alcohol fuel program has been implemented. In 1997,  
21 approximately 4 million automobiles ran on neat ethanol and approximately nine million  
22 automobiles ran on a 22 percent ethanol-blended gasoline mixture (Grosjean et al.,  
23 2002a). However, data from Brazil are limited by the lack of ambient monitoring data

1 prior to ethanol fuel use. Nonetheless, it is clear from data in Brazil that significant  
2 increases in ambient acetaldehyde and PAN (which can be formed by photooxidation of  
3 acetaldehyde) resulted from greater emissions of acetaldehyde and ethanol associated  
4 with increased ethanol use (Andrade et al., 1998; Montero et al, 2001; Colon et al, 2001;  
5 Grosjean et al., 2002b; Martins and Arbilla, 2003).

6  
7 Monitoring and modeling studies of ethanol impacts resulting from U. S. winter  
8 oxygenated fuel programs can also provide insights. A study in Albuquerque, New  
9 Mexico, where 10 percent ethanol was used in a winter oxygenated fuel program, found  
10 elevated levels of acetaldehyde and PAN (Gaffney et al., 1997). However, a similar  
11 study in Denver, Colorado, found no elevation of acetaldehyde (Anderson, et al., 1994,  
12 1996, 1997). One explanation hypothesized that photochemical production and  
13 destruction of these compounds suppressed the emissions effect. Alkenes in particular  
14 play a large role in acetaldehyde production (Altshuller 1991a, 1991b, 1993; Calvert et al.,  
15 2000). Furthermore, a modeling study in California found large increases in ethanol  
16 concentration, but relatively small impacts on acetaldehyde with ten percent ethanol use  
17 in winter (Allen et al., 1999).

18  
19 Given that currently available data are only available from modeling and  
20 monitoring studies with very different conditions than those likely to occur in the United  
21 States under EISA, an assessment of likely impacts for a realistic scenario is needed. In  
22 this paper, we present a comprehensive assessment of potential emission inventory and  
23 air quality impacts that could result in 2022 with full implementation of requirements

1 under EISA, considering emission changes in all sources across the fuel lifecycle. This is  
2 the first time that a full, 3-D modeling study has been performed using realistic future  
3 emission scenarios and a full upstream and downstream inventory. This assessment  
4 includes inventories for the following pollutants: VOCs, NO<sub>x</sub>, CO, particulate matter  
5 microns in diameter and less (PM<sub>10</sub>), particulate matter 2.5 microns in diameter and less  
6 (PM<sub>2.5</sub>), sulfur dioxide (SO<sub>2</sub>), ammonia (NH<sub>3</sub>), benzene, 1,3-butadiene, acrolein,  
7 formaldehyde, acetaldehyde, and ethanol. Air quality modeling results are presented for  
8 ozone and air toxics. While PM<sub>2.5</sub> concentrations will also be impacted, and these  
9 impacts have been modeled (U. S. EPA, 2010a), air quality modeling results for this  
10 pollutant are not discussed in this paper because of limitations in local-scale results,  
11 resulting from an error in spatially allocating emissions from one PM<sub>2.5</sub> emission source.  
12 We compared modeled emissions and air quality under EISA to a base case scenario  
13 which assumed renewable fuel volumes at the level mandated by U. S. EPA's 2007  
14 Renewable Fuel Standard (RFS1; U. S. DOE, 2007; U. S. EPA, 2007b).

15

## 16 **2. Methodology**

17

18 Modeling was done for calendar year 2022, for two scenarios. The first was a  
19 baseline scenario which assumed a volume of 7.5 billion gallons of renewable fuels, as  
20 mandated by the U. S. EPA's 2007 renewable fuel standard (RFS1) rule (U. S. EPA,  
21 2007b). The second scenario included impacts of EISA, which was implemented by the  
22 RFS2 rule (U. S. EPA, 2010a). Based on EISA requirements and an U. S. EPA  
23 assessment of production feasibility (U. S. EPA, 2009a), this scenario assumed that the

1 renewable fuel mandate of 36 billion gallons would be reached with 15 billion gallons of  
2 corn ethanol, 16 billion gallons of cellulosic ethanol, and 3 billion gallons of imported  
3 ethanol. The remainder of required renewable fuel volumes would be met with  
4 renewable diesel and biodiesel fuel. Approximately 20 billion gallons of the ethanol will  
5 be in the form of E85. U. S. EPA's final RFS2 rule implementing requirements of EISA  
6 assumed that requirements would be met with lower volumes of cellulosic ethanol and  
7 higher volumes of cellulosic diesel fuel (U. S. EPA, 2010a).

8

### 9 *2.1. Emission Inventory*

10

11 State-level light-duty highway vehicle exhaust emissions for all pollutants except  
12 ethanol were estimated with a draft version of the Motor Vehicle Emissions Simulator  
13 (MOVES) model (U. S. EPA, 2009b). MOVES contains significant updates to U. S.  
14 EPA's previous highway vehicle emissions model, MOBILE6. Fuel properties used as  
15 model inputs were based on data from refinery modeling to account for how gasoline  
16 properties would be likely to change with increased use of ethanol. In modeling impacts  
17 of RFS2, ethanol was assumed to be blended at either 10 percent (E10) or 85 percent  
18 (E85) with gasoline, with no intermediate level blends. It must be emphasized that there  
19 were significant limitations in emissions data quantifying the effects of E10 on advanced  
20 technology vehicle emissions and the effects of E85 on all vehicles (U. S. EPA, 2010a).  
21 U. S. EPA relied on unpublished data from a test program then in progress to quantify  
22 E10 impacts; these data are discussed in U. S. EPA (2010a; pp. 520-521). Based on  
23 these limited data, E10 use was assumed to decrease VOC emission rates by 7-10% and

1 increase NO<sub>x</sub> emissions by over 7%. However, some recent test data were not consistent  
2 with these assumptions, and more data are needed to draw firm conclusions. Furthermore,  
3 vehicles using E85 were assumed to have lower emissions of PM and CO, but this  
4 assumption is also based on limited data (Southwest Research Institute, 2007;  
5 Environment Canada, 2007; Graham et al., 2008). No reductions in VOC or NO<sub>x</sub>  
6 emissions were assumed for E85. Two studies (Environment Canada, 2007; Graham et  
7 al., 2008) showed significant NO<sub>x</sub> reductions with E85, but data from EPA's Certification  
8 and Fuel Economy System showed a small impact. Overall, effects were not statistically  
9 significant due to a large amount of variability in the data (U. S. EPA, 2010a). In  
10 contrast to this modeling effort, Jacobson (2007) assumed significant NO<sub>x</sub> effects with  
11 E85. Also, E85 emissions were estimated in MOVES by applying multiplicative  
12 adjustments to E0 emissions. These multiplicative adjustments did not vary with  
13 temperature. Thus, emissions of vehicles running on E85 were assumed to change  
14 proportionally to emissions of vehicles running on E0 in response to temperature. State-  
15 level emissions from MOVES were allocated to counties using data from U. S. EPA's  
16 National Mobile Inventory Model, v.2008 (NMIM2008; U. S. EPA, 2009c). A cost  
17 effectiveness analysis was done to determine where E85 profit margins would be highest,  
18 and higher levels of E85 use were assumed to occur in these areas. These areas of higher  
19 E85 use include New England, much of the Midwest, the Pacific Northwest, and  
20 Southern California (U. S. EPA, 2009a).

21

22 Light-duty vehicle evaporative, heavy-duty vehicle, motorcycle, and nonroad  
23 equipment emissions, except for ethanol, were estimated at the county level using

1 NMIM2008 (U. S. EPA, 2009c). Stationary source emissions were obtained by  
2 procedures describe in the U. S. EPA Office of Air Quality Planning and Standards, 2005  
3 Modeling Platform documentation (<http://www.epa.gov/ttn/chief/emch/index.html>). In  
4 this platform, year 2022 projections were made from a year 2005 baseline. Impacts of  
5 RFS2 on stationary source emissions, except for ethanol, were estimated using a U. S.  
6 EPA spreadsheet model based on the GREET (Greenhouse Gases, Regulated Emissions,  
7 and Energy Use in Transportation) model, originally developed by the U. S. Department  
8 of Energy’s Argonne National Laboratory (U. S. DOE, 2009, U. S. EPA, 2010a).  
9 Improvements to GREET included a significant expansion of modeling agricultural  
10 sector impacts, employing economic and agriculture models to consider factors such as  
11 land-use impact, agricultural burning, fertilizer, pesticide use, livestock, crop allocation,  
12 and crop exports. Other updates and enhancements included updated emission factors for  
13 fuel and feedstock transport. In addition, the most recent version of GREET did not  
14 include air toxics; thus, emission factors for benzene, 1,3-butadiene, acrolein,  
15 formaldehyde, and acetaldehyde were added.

16

17 *2.2. Air Quality Modeling*

18

19 Air quality modeling was conducted for the 48 contiguous states, using the  
20 Community Multi-scale Air Quality Model (CMAQ), version 4.7, with the Carbon Bond  
21 05 (CB05) chemical mechanism (Byun and Schere, 2006, <http://www.cmaq-model.org>).  
22 The modeling domain was made up of a large continental U.S. 36 kilometer (km) grid  
23 and two 12 km grids (an Eastern U.S. and a Western U.S. domain), as shown in Figure 1.

1 The conditions from the 36 km coarse grid modeling were used as the initial/boundary  
2 state for subsequent 12 km finer grid modeling. Since 12 km domains overlap, data from  
3 the 12-km eastern domain data were used for all states from North Dakota to Texas  
4 (except El Paso, TX) and east and data from the the 12-km western domain were used for  
5 all states from Montana to New Mexico along with El Paso, TX, and west. A 12 km grid  
6 size rather than a coarser grid was used for modeling, since finer grids are able to capture  
7 variations in both VOC and NO<sub>x</sub> emissions and their effects on nonlinearities in ozone  
8 photochemistry (Schere, 1988, Cohan et al., 2006). The modeling domain contained 14  
9 vertical layers with the top of the modeling domain at about 16 km. The hourly results  
10 were processed into daily maximum 8-hour ozone concentrations over the ozone season  
11 (June-September), as well as seasonal and annual averages for air toxics.

12

13 Emission inventories were prepared for input into CMAQ using the SMOKE  
14 (Sparse Matrix Operator Kernel Emissions) emissions processing system  
15 (<http://www.smoke-model.org>). SMOKE allocates emissions spatially and temporally,  
16 using spatial allocation surrogates to allocate area and mobile source emissions to grid  
17 cells and temporal activity profiles to allocate emissions to hour of day. Depending on  
18 the source, spatial surrogates are based on types of land use, census data, employment  
19 information, or other factors. SMOKE also speciates hydrocarbon and particulate matter  
20 emissions into individual chemical compounds using a comprehensive set of speciation  
21 profiles. Ethanol emissions were estimated in this speciation step.

22

1           While in general, county-level emissions of area and mobile sources were  
2 spatially allocated to grid cells using surrogates from the SMOKE model, there were  
3 some exceptions. Since increased use of ethanol results in higher emissions associated  
4 with production and distribution, approaches had to be developed to allocate these  
5 increased emissions to locations where plants were likely to be located. Increased corn  
6 ethanol production was expected to occur at existing or planned plants, but likely  
7 locations for cellulosic ethanol plants and associated activity were unknown. Emissions  
8 from these plants were treated as area source emissions and allocated to counties based  
9 on potential cellulosic feedstock sites across the U.S. that could economically justify the  
10 construction of a cellulosic plant facility (U. S. EPA, 2010a). Emissions associated with  
11 distribution were allocated based on an analysis by Oak Ridge National Research  
12 Laboratory of projected ethanol transport modes, distances, and volumes transferred  
13 under various ethanol volume scenarios (U. S. EPA, 2010a).

14  
15           The CMAQ simulations were done for the entire year of 2005, using  
16 meteorological inputs developed by U. S. EPA. The meteorological inputs for air quality  
17 modeling were derived from an offline simulation of the Pennsylvania State  
18 University/National Center for Atmospheric Research Mesoscale Model, MM5 (Grell et  
19 al., 1994). MM5 uses a limited-area, nonhydrostatic, terrain-following coordinate system  
20 that solves the full set of physical and thermodynamic equations which govern  
21 atmospheric motions. In this application, 34 vertical layers were used and the three-  
22 dimensional analyses of temperature, moisture and wind field were nudged towards  
23 observations. The simulated meteorological fields were found to closely match observed

1 synoptic patters and to compare well to observed values of meteorological parameters (U.  
2 S. EPA, 2010b). The meteorological outputs from MM5 were processed to create model-  
3 ready inputs for CMAQ using the Meteorology-Chemistry Interface Processor (MCIP)  
4 version 3.4 (U. S. EPA, 1999). Boundary and initial species concentrations were  
5 provided by a three-dimensional global atmospheric chemistry model, the GEOS-CHEM  
6 model, run for 2005 with a grid resolution of 2.0 x 2.5 degrees (latitude-longitude) and 30  
7 vertical layers (Le Sager et al., 2008).

8  
9 In addition to modeling 2022 scenarios, modeling was done for calendar year  
10 2005, for purposes of conducting an operational model performance evaluation using  
11 2005 air quality monitoring data. An extensive discussion of this performance evaluation  
12 can be found in U. S. EPA (2010b). The comparison included statistical assessment of  
13 modeled versus monitored data paired in space and time. Results of this evaluation  
14 indicated that performance was similar to that found in other recent U. S. EPA  
15 applications, indicating that application of CMAQ using this modeling platform provides  
16 a scientifically credible approach for assessing the impacts of future emissions changes  
17 on air quality. However model performance for ozone, benzene, formaldehyde and  
18 acetaldehyde was better than performance for 1,3-butadiene and acrolein, where there  
19 was large bias and error. For butadiene, the approximate bias at the national level was  
20 about -65% and the error was about 85%. For acrolein, the bias was about -90% with a  
21 100% error. The bias and error may be due to several causes; chief among them are  
22 emissions errors and ambient measurement errors. As a result, findings for 1,3-butadiene  
23 and acrolein should be viewed as more uncertain.

1

2 **3. Results**

3

4 *3.1. Nationwide Inventory Impacts*

5

6 Table 1 presents 48 state emission inventories assuming renewable fuel volumes  
7 mandated by RFS1 versus RFS2 in 2022, and the percent change in these emissions  
8 under RFS2 relative to RFS1. Emissions are broken down into contributions from  
9 upstream and downstream sources in the supplementary information, Table S1. Carbon  
10 monoxide, benzene, and 1,3-butadiene are projected to decrease in 2022 with RFS2  
11 renewable fuel volumes, while NO<sub>x</sub>, hydrocarbons (HC) and the other air toxics,  
12 especially acetaldehyde, are projected to increase due to the impacts of ethanol. The  
13 large estimated decrease in CO is due primarily to impacts of ethanol on exhaust  
14 emissions from vehicles running on E10 and E85 and nonroad equipment running on E10.  
15 The decrease in benzene is due primarily to displacement of gasoline with ethanol in the  
16 fuel pool. Increases in NO<sub>x</sub> result from a combination of increases in vehicle and  
17 nonroad equipment emissions due to higher volumes of E10 gasoline, and increased  
18 emissions from corn and cellulosic ethanol production. Small projected increases in  
19 hydrocarbons are due to increases in vehicle and nonroad equipment emissions running  
20 on E10, as well as emissions connected with renewable fuel production (countered by  
21 decreases in emissions associated with gasoline production and distribution as ethanol  
22 displaces some of the gasoline). Increases in SO<sub>x</sub> emissions are due to increases in  
23 agricultural chemical production and transport, while substantial PM increases are also

1 associated with fugitive dust from agricultural operations. Ammonia emissions are  
2 expected to increase slightly due to increased ammonia from fertilizer use. While PM  
3 emissions from vehicles decrease as a result of increased E85 volumes, there are  
4 localized increases associated with fugitive dust from agricultural operations, as well as  
5 ethanol production and transport. Emissions of ethanol and acetaldehyde increase  
6 substantially as a result of emissions from vehicles and engines and increased emissions  
7 from ethanol production and transport.

8

### 9 *3.2. Air Quality Impacts*

10

#### 11 *3.2.1. Ozone*

12

13 Figure 2 presents changes in 8-hour ozone design values in 2022 between the  
14 EISA scenario and the RFS1 scenario. An ozone design value is defined as the three-  
15 year average of the annual, fourth-highest daily maximum 8-hour average ozone  
16 concentration at a given monitor (Code of Federal Regulations, 40 CFR, Part 50,  
17 Appendix P). To predict 8-hour ozone design values the CMAQ predictions were used in  
18 a relative sense by combining the 2005 base year predictions with predictions from the  
19 2022 scenarios and applying these modeled ratios (2022/2005) to existing ambient air  
20 quality observations. To account for interannual variability in observed ozone levels, the  
21 ambient air quality observations used here were an average of the three ozone design  
22 values centered around the 2005 base year (i.e., 2003-2005, 2004-2006, 2005-2007).  
23 Using this standard air quality modeling methodology (U. S. EPA, 2007c), it is projected

1 that achieving the required renewable fuel volumes by 2022 would adversely impact  
2 ozone air quality over much of the U.S. However, ozone air quality improvements are  
3 projected in a few highly-populated areas which currently have poor air quality.  
4

5 As can be seen in Figure 2, most of the counties where impact on design value  
6 was calculated, especially those in the Midwest, see increases in their ozone design  
7 values. The majority of these design value increases are more than 0.2 ppb but less than  
8 0.5 ppb. However, there are some counties that will see 8-hour ozone design value  
9 increases above 0.5 ppb; these counties are mainly in the eastern U.S. There are also  
10 some counties that are projected to see 8-hour ozone design value decreases. The  
11 counties with ozone design value decreases greater than 0.5 ppb are in Southern  
12 California.  
13

14 Table 2 shows the average change in 2022 8-hour ozone design values for: (1) all  
15 counties with 2005 baseline design values, (2) counties with 2005 baseline design values  
16 that exceeded the 2008 ozone standard, (3) counties with 2005 baseline design values that  
17 did not exceed the 2008 standard but were within ten percent of it, (4) counties with  
18 2022 design values that exceeded the 2008 ozone standard, and (5) counties with 2022  
19 design values that did not exceed the standard but were within ten percent of it. Many of  
20 these statistics show an increase in ozone design values in 2022, but the magnitude of the  
21 increase varies and there are some statistics which show a decrease in 8-hour ozone  
22 design values as a result of increased ethanol usage.  
23

1           The projected ozone increases in some areas and decreases in other areas are  
2 likely a result of the emissions changes due to the increased volumes of renewable fuels  
3 combined with the photochemistry involved, differing emission rates of VOCs and NO<sub>x</sub>  
4 in different areas of the country, and the meteorological conditions in different areas of  
5 the country. When VOC levels are high relative to NO<sub>x</sub> (e.g., in areas with large  
6 quantities of biogenic VOC emissions), ozone is reduced by NO<sub>x</sub> reductions and  
7 increased by NO<sub>x</sub> increases. Such conditions are called “NO<sub>x</sub>-limited” or “NO<sub>x</sub>-  
8 sensitive.” The ozone increases seen in the southeastern U.S. and many of the other rural  
9 areas are likely due to the fact that those areas are NO<sub>x</sub>-limited and the changes being  
10 modeled are projected to increase NO<sub>x</sub>. However, when NO<sub>x</sub> levels are relatively high  
11 and VOC levels relatively low, increases in NO<sub>x</sub> can decrease ozone, because NO reacts  
12 directly with ozone and NO<sub>2</sub> terminates radicals, forming nitric acid, which is removed  
13 from the system or can form particulate nitrates. Such conditions are called “VOC-  
14 limited” or “NO<sub>x</sub>-saturated.” Under these conditions, VOC reductions are effective in  
15 reducing ozone, but NO<sub>x</sub> reductions can actually increase local ozone under certain  
16 circumstances. The ozone decreases seen in southern California and some of the other  
17 urban areas, like Cleveland and Miami, are likely due to the fact that these are VOC-  
18 limited areas and they are projected to experience increases in NO<sub>x</sub> due to RFS2  
19 requirements, both of which would result in ozone decreases. While RFS2 is projected to  
20 lower ozone levels in southern California under our current assumptions of 2022  
21 conditions, it is important to recognize that the State has plans (CARB, 2007) to  
22 substantially lower NO<sub>x</sub> emissions in the future which may bring this area into a NO<sub>x</sub>-  
23 limited regime. In that context, the NO<sub>x</sub> increases here run counter to State efforts and

1 may lead to ozone increases if the planned NO<sub>x</sub> reductions are achieved sometime in the  
2 future.

3

4 Incremental impacts of emissions from vehicles running on E85 fuel in the RFS2  
5 scenario can be made by comparing RFS2 results to an alternative baseline scenario  
6 which consists largely of E10. Results of such a comparison are presented in the  
7 supplementary information. The alternative baseline scenario assumed ethanol volumes  
8 projected by the Department of Energy's Annual Energy Outlook (AEO) for 2007  
9 (Energy Information Administration, 2007). Ethanol volumes for this alternative baseline  
10 scenario relative to the RFS1 and RFS2 cases are provided in Table S2, and inventory  
11 impacts in Table S3. Table S4 compares changes in 8-hour ozone design values for  
12 RFS2 relative to the two baselines, and Figure S-1 depicts changes in design values for  
13 RFS2 relative to the alternative baseline. These results suggest that incremental impact  
14 of E85 use is to increase ozone design values with a similar spatial pattern of increases  
15 and decreases as the comparison of RFS2 results to the RFS1 baseline.

16

### 17 3.2.2. *Air Toxics*

18

19 Results of this assessment indicate that, while there are some localized impacts,  
20 the renewable fuel volumes required by RFS2 have relatively little impact on national  
21 average ambient concentrations of the modeled air toxics. An exception is increased  
22 ambient concentrations of ethanol.

23

1           The relatively small impact was particularly surprising for acetaldehyde, given the  
2 increase in acetaldehyde emissions in Table 1. Annual percent changes in ambient  
3 concentrations of acetaldehyde are less than one percent for most of the country (Figure  
4 3). Several urban areas show decreases in ambient acetaldehyde concentrations ranging  
5 from 1-10%, and some rural areas associated with new ethanol plants show increases in  
6 ambient acetaldehyde concentrations ranging from 1-10% with the fuel volumes required  
7 by RFS2. Annual absolute changes in ambient concentrations of acetaldehyde are  
8 generally less than 0.1  $\mu\text{g}/\text{m}^3$ . The largest increases in ambient acetaldehyde  
9 concentrations with RFS2 volumes occur in areas associated with new ethanol plants.  
10 This result is due to an increase in emissions of primary acetaldehyde and precursor  
11 emissions from ethanol plants not included in the RFS1 baseline scenario. The relatively  
12 small overall impact on acetaldehyde concentrations is likely attributable to the  
13 complexity of the chemical formation of acetaldehyde. Most ambient acetaldehyde is  
14 formed from secondary photochemical reactions of numerous precursor compounds, and  
15 many photochemical mechanisms are responsible for this process. As discussed  
16 previously, reductions in certain acetaldehyde precursors, primarily alkenes, may offset  
17 the impacts of increases in direct emissions, and the acetaldehyde produced from  
18 photochemical transformation of increased ethanol emissions. Analysis of data inputs  
19 used in this assessment revealed a significant decrease in alkene levels estimated from  
20 speciated emission profiles for ten percent ethanol gasoline versus non-ethanol gasoline;  
21 these speciation profiles, which are used to break total hydrocarbons down to individual  
22 constituent compounds, were applied to vapor losses associated with storage and  
23 transport of gasoline. However, the profiles used may not be representative of real-world

1 conditions (U. S. EPA 2010a). A limited sensitivity analysis with more representative  
2 profiles developed after modeling was completed, using newly collected data, suggested  
3 acetaldehyde increases in many areas where decreases were projected.

4

5 Modeling results do not show substantial impacts on ambient concentrations of  
6 formaldehyde. As shown in Figure 4, most of the U.S. experiences a one percent or less  
7 change. Decreases in ambient formaldehyde concentrations range between one and five  
8 percent in a few urban areas. Increases range between 1-2.5% in some rural areas  
9 associated with new ethanol plants; this result is due to increases in emissions of primary  
10 formaldehyde and formaldehyde precursors from the new ethanol plants.

11

12 Modeling results suggest that the renewable fuel volumes required by the RFS2  
13 rule will lead to small nationwide decreases in ambient benzene concentrations. As  
14 shown in Figure 5, decreases in ambient benzene concentrations range between 1-10%  
15 across most of the country and can be higher in a few urban areas. Absolute changes in  
16 ambient concentrations of benzene are all less than 0.2  $\mu\text{g}/\text{m}^3$ . The modeling also shows  
17 small increases and decreases in ambient concentrations of 1,3-butadiene in parts of the  
18 U.S (Figure 6). Generally, decreases occur in some southern areas of the country and  
19 increases occur in some northern areas and areas with high altitudes. Changes in absolute  
20 concentrations of ambient 1,3-butadiene are generally less than 0.005  $\mu\text{g}/\text{m}^3$ . The largest  
21 1,3-butadiene increases appear in rural areas with cold winters and low ambient levels but  
22 high contributions of emissions from snowmobiles. However, the snowmobile emission  
23 factor data that were used to develop inventories were obtained from only three engines

1 and showed significant increases in 1,3-butadiene emissions with ten percent ethanol  
2 gasoline; it is likely this increase is not representative of the fleet as a whole. Small  
3 increases and decreases are also seen for acrolein (Figure 7). Decreases in acrolein  
4 concentrations occur in some eastern and southern parts of the U.S., and increases occur  
5 in some northern areas (due to 1,3-butadiene emissions from snowmobiles, because 1,3-  
6 butadiene is an acrolein precursor) and areas associated with new ethanol plants.

7  
8 As expected, the renewable fuel volumes required by RFS2 will lead to  
9 significant nationwide increases in ambient ethanol concentrations. Increases ranging  
10 between 10 to 50 percent are seen across most of the country (Figure 8). The largest  
11 increases (more than 100 percent) occur in urban areas with high amounts of onroad  
12 emissions and in rural areas associated with new ethanol plants. Absolute increases in  
13 ambient ethanol concentrations are above 1.0 ppb in some urban areas. However,  
14 sensitivity tests using alternative profiles show that ethanol impacts could be reduced by  
15 more than ten percent across much of the modeling domain (U. S. EPA, 2010b).

#### 16 17 **4. Summary and Discussion**

18  
19 While this assessment models realistic conditions which could result from  
20 implementation of renewable fuel standards required by RFS2, actual levels of renewable  
21 fuels are likely to be different from those modeled. Because increased use of ethanol  
22 impacts emissions at many different points in the fuel life-cycle, the direction and  
23 magnitude of air quality impacts for ozone and air toxics with the renewable fuel mandate

1 of the RFS2 rule will likely vary across the country. Results of this study support  
2 concerns that increased use of ethanol may adversely impact ozone in many parts of the  
3 United States, including many nonattainment areas, although some urban areas will see  
4 reductions. In general, the ozone results are consistent with the findings of Jacobson  
5 (2007). Study results do not suggest large impacts on air toxic concentrations across the  
6 country, although there may be localized increases in some areas, particularly those  
7 associated with fuel production. While increased use of ethanol will certainly result in  
8 significantly increased emissions of acetaldehyde, these results do not show a significant  
9 impact on ambient concentrations, due to reductions in acetaldehyde precursor emissions.  
10 This result may seem to be inconsistent with some other studies, such as the Jacobson  
11 (2007) study and monitoring studies in Brazil; however, these other studies involved  
12 much higher levels of ethanol, and in the case of the Brazil studies, conditions not  
13 relevant to the U. S. Also, while Jacobson (2007) showed increases in acetaldehyde and  
14 formaldehyde since he modeled an extreme case of 100% penetration of E85, these  
15 increases were offset by reductions in benzene and 1,3-butadiene. When changes in  
16 population-weighted mixing ratios were weighted by cancer unit risk estimates, there was  
17 little net impact on overall cancer risk, and Jacobson concluded that enhanced ozone risk  
18 was a greater concern than air toxics with increased ethanol use. The results of this study  
19 support that conclusion.

20

21 It should be emphasized that these results must be interpreted with the  
22 understanding that there are considerable limitations and uncertainties in inventories,  
23 atmospheric processes in CMAQ, and other aspects of the modeling process. A

1 discussion of these limitations and uncertainties can help elucidate directions for research  
2 needed to better characterize potential air quality impacts. First, modeling assumed that  
3 ethanol will make up 34 of the 36 billion gallon renewable fuel mandate, and that  
4 approximately 20 billion gallons of this ethanol will be in the form of E85. The emission  
5 impacts and air quality results would be different if, instead of E85, more non-ethanol  
6 renewable fuels are used or mid-level ethanol blends are approved. In fact, U. S. EPA's  
7 final RFS2 rule assumed E85 volumes are likely to be significantly lower than what we  
8 assumed for this air quality assessment (U. S. EPA, 2010a). Assumptions about the types  
9 of renewable fuels and ethanol blend levels are one of the largest sources of uncertainties  
10 in this analysis.

11

12 A second major source of uncertainty is limited emissions data for vehicles  
13 running on E85, and for advanced technology vehicles running on lower level ethanol  
14 blends. Thus estimated impacts on NO<sub>x</sub>, VOC, and air toxics have considerable  
15 uncertainty. As mentioned previously, emissions of vehicles running on E85 were  
16 assumed to change proportionally to emissions of vehicles running on E0 in response to  
17 temperature. However, if this assumption is not valid, as suggested by some limited  
18 emissions data, modeled ozone formation at lower temperatures could be impacted  
19 (Ginnebaugh et al., 2010).

20

21 In addition, assumptions made about impacts of ethanol on NO<sub>x</sub> emissions may  
22 have a significant impact on modeling results. This modeling assumes increases in NO<sub>x</sub>  
23 emissions with E10 and no change with E85. Previous EPA modeling analyses have

1 shown that reducing NO<sub>x</sub> emissions will have widespread ozone benefits (U. S. EPA,  
2 2008). It was not within the scope of this analysis to conduct air quality modeling  
3 simulations of various ethanol/NO<sub>x</sub> combinations, but it is expected that the ozone  
4 response in some locations is highly sensitive to these assumptions.

5  
6 Clearly, emission testing of advanced technology vehicles on a variety of ethanol  
7 blends is needed to lower the uncertainty associated with this analysis. Other sources of  
8 uncertainty are estimates of emissions from ethanol plants, particularly cellulosic ethanol  
9 plants, and emissions associated with storage and transport of fuel. All of these estimates  
10 are based on very limited data. As mentioned previously, there is no way of knowing  
11 where future plants will be located, and siting is based on economic assumptions.  
12 Moreover, there are numerous assumptions about land use changes that impact  
13 inventories for upstream sources and consequently can impact air quality modeling  
14 results. In addition to assumptions that affect emission inventories, limitations in data  
15 used to develop hydrocarbon speciation profiles introduce uncertainty into modeling  
16 results, as discussed previously. We are currently collecting updated emission profiles  
17 for different biofuel mixtures, based on ongoing measurements with new engines under  
18 varied driving cycles. With these profiles we will test the sensitivity of the ozone, PM<sub>2.5</sub>  
19 and HAPs results in this paper to variations in VOC profiles and NO<sub>x</sub> uncertainties.

20  
21 Another lesser source of uncertainty is the photochemical mechanisms used in the  
22 air quality model. Ozone is formed entirely through thousands of photochemical  
23 reactions involving all emitted VOCs and NO<sub>x</sub> and its representation in air quality models

1 must be highly condensed. Other pollutants such as acetaldehyde, formaldehyde, and  
2 acrolein have substantial contributions from atmospheric chemical reactions.  
3 Formaldehyde and acetaldehyde are produced as byproducts of almost every VOC that  
4 reacts. Since secondarily formed pollutants can result from many different reaction  
5 pathways, they are affected by uncertainties associated with any pathway, as well as  
6 uncertainties in the emissions of their precursors.

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## 13 14 **Disclaimer**

15  
16 This paper has been reviewed in accordance with U. S. EPA peer and  
17 administrative review policies and approved for presentation and publication. This work  
18 does not necessarily reflect U. S. EPA policies or views. Mention of trade names or  
19 commercial products does not constitute endorsement or recommendation for their use.

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1 **Table 1**

2 Nationwide emission inventories for 2022, RFS2 versus RFS1.

3

Pollutant	US Total RFS1	US Total RFS2	RFS2 vs RFS1
	Annual Short Tons	Annual Short Tons	Percent Change
NOx	11,415,147	11,781,115	3.21%
HC	10,292,785	10,412,658	1.16%
PM10	11,999,983	12,068,629	0.57%
PM2.5	3,371,024	3,389,223	0.54%
CO	51,631,075	47,011,171	-8.95%
Benzene	226,683	217,021	-4.26%
1,3-Butadiene	14,458	14,264	-1.34%
Acetaldehyde	58,405	65,722	12.53%
Formaldehyde	140,156	140,330	0.12%
Acrolein	6,399	6,477	1.23%
Ethanol	457,071	906,719	98.37%
SO2	8,878,706	8,936,086	0.65%
NH3	4,213,048	4,213,189	0.00%

4

5

1 **Table 2.**

2 Average Change in Projected Future Year 8-hour Ozone Design Value, RFS2 versus

3 RFS1.

4

AVERAGE <sup>a</sup>	NUMBER OF US COUNTIES	2020 POPULATION <sup>b</sup>	CHANGE IN 2022 DESIGN VALUE (PPB) RFS2-RFS1
All	678	238,378,000	0.46
All, population-weighted	678	238,378,000	0.28
Counties whose 2005 base year is violating the 2008 8-hour ozone standard	389	174,967,000	0.44
Counties whose 2005 base year is violating the 2008 8-hour ozone standard, population-weighted	389	174,967,000	0.26
Counties whose 2005 base year is within 10 percent of the 2008 8-hour ozone standard	208	43,172,000	0.52
Counties whose 2005 base year is within 10 percent of the 2008 8-hour ozone standard, population-weighted	208	43,172,000	0.35
Counties whose 2022 RFS2 control case is violating the 2008 8-hour ozone standard	26	41,017,000	0.04
Counties whose 2022 RFS2 control case is violating the 2008 8-hour ozone standard, population-weighted	26	41,017,000	-0.14
Counties whose 2022 RFS2 control case is within 10% of the 2008 8-hour ozone standard	110	61,618,000	0.34
Counties whose 2022 RFS2 control case is within 10% of the 2008 8-hour ozone standard, population-weighted	110	61,618,000	0.31

5

Notes:

6

<sup>a</sup> Changes are only calculated for counties with a valid base year design value

7

<sup>b</sup> Woods & Poole Economics, Inc. (2001). Population by Single Year of Age CD.

8

1 Fig. 1. Map of the CMAQ modeling domain.

2

3 Fig. 2. Changes in 2022 8-hour ozone design values for nonattainment areas between the  
4 RFS2 scenario and the RFS1 scenario.

5

6 Fig. 3. Annual percent changes in ambient concentrations of acetaldehyde between the  
7 RFS2 scenario and the RFS1 scenario.

8

9 Figure 4. Annual percent changes in ambient concentrations of formaldehyde between  
10 the RFS2 scenario and the RFS1 scenario.

11

12 Figure 5. Annual percent changes in ambient concentrations of benzene between the  
13 RFS2 scenario and the RFS1 scenario.

14

15 Figure 6. Annual percent changes in ambient concentrations of 1,3-butadiene between  
16 the RFS2 scenario and the RFS1 scenario.

17

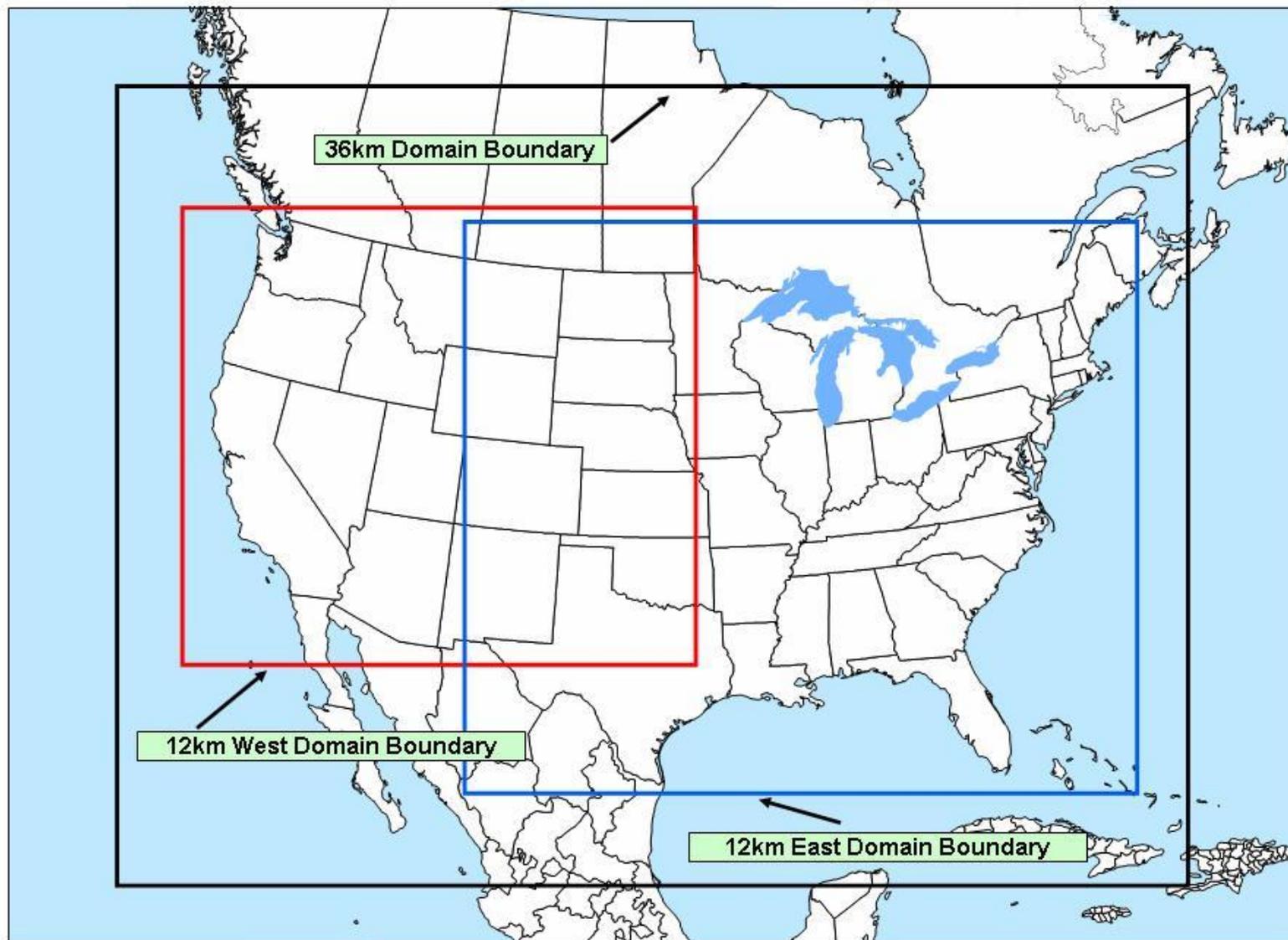
18 Figure 7. Annual percent changes in ambient concentrations of acrolein between the  
19 RFS2 scenario and the RFS1 scenario.

20

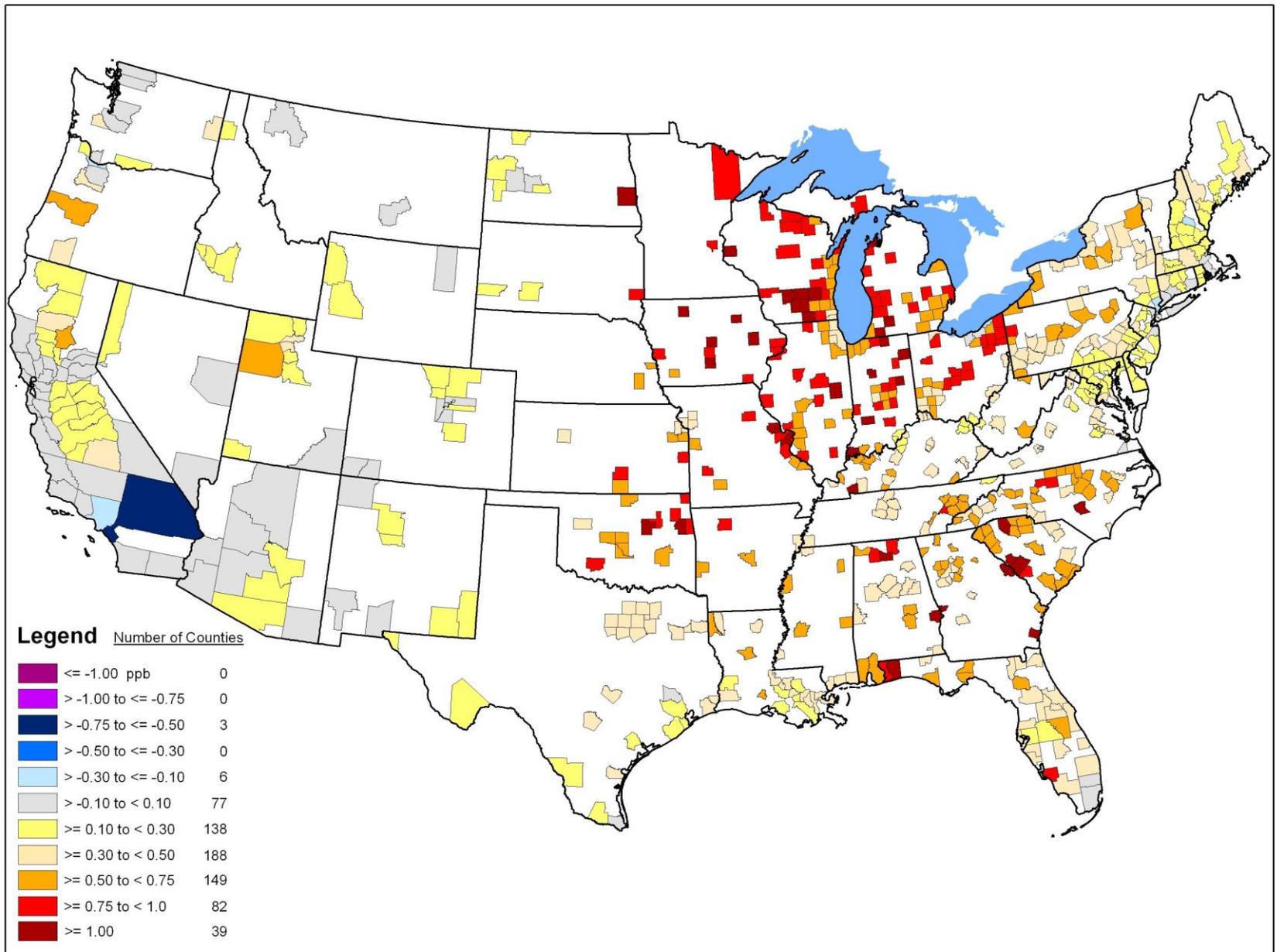
21 Figure 8. Annual percent changes in ambient concentrations of ethanol between the  
22 RFS2 scenario and the RFS1 scenario.

23

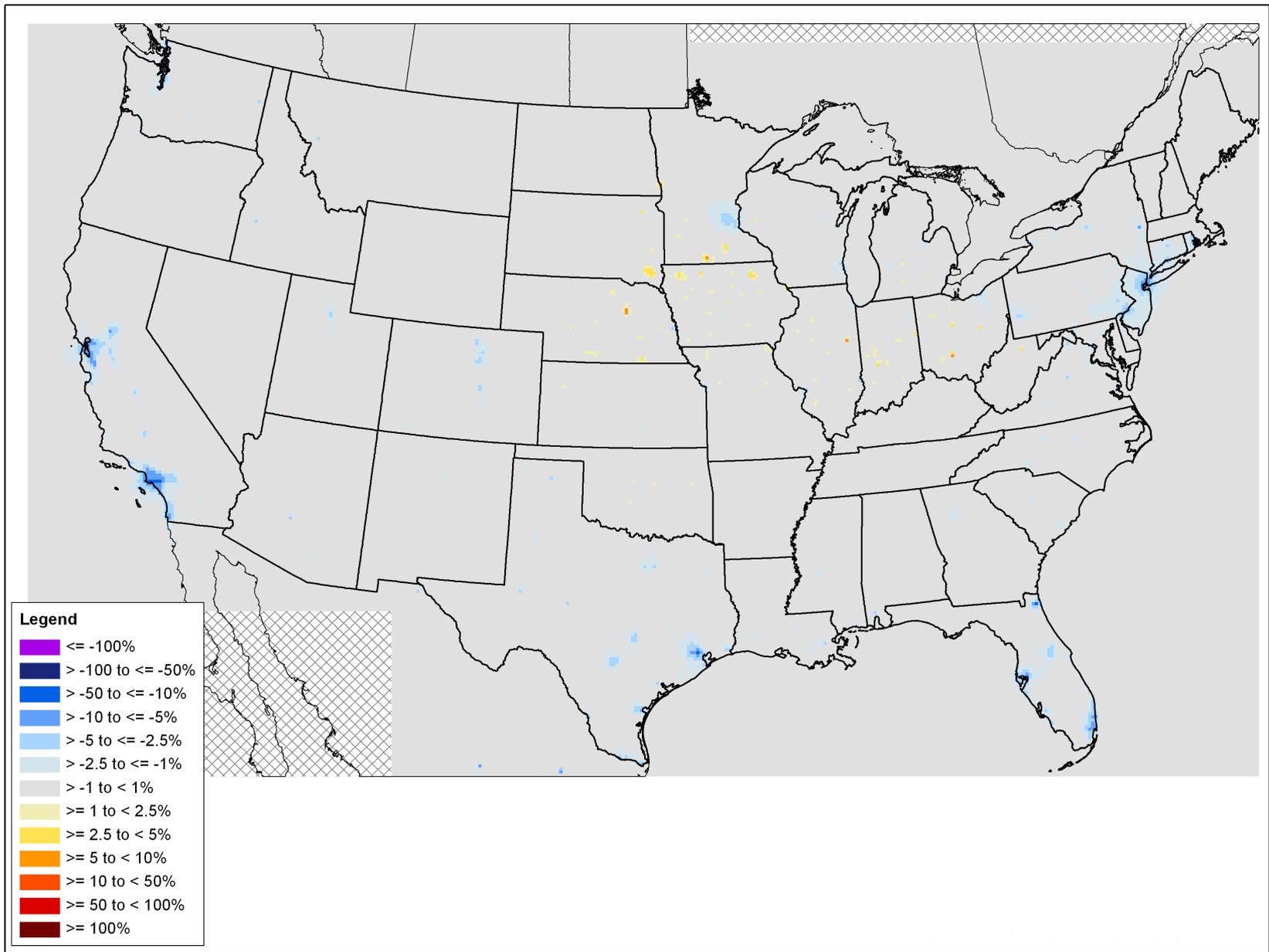
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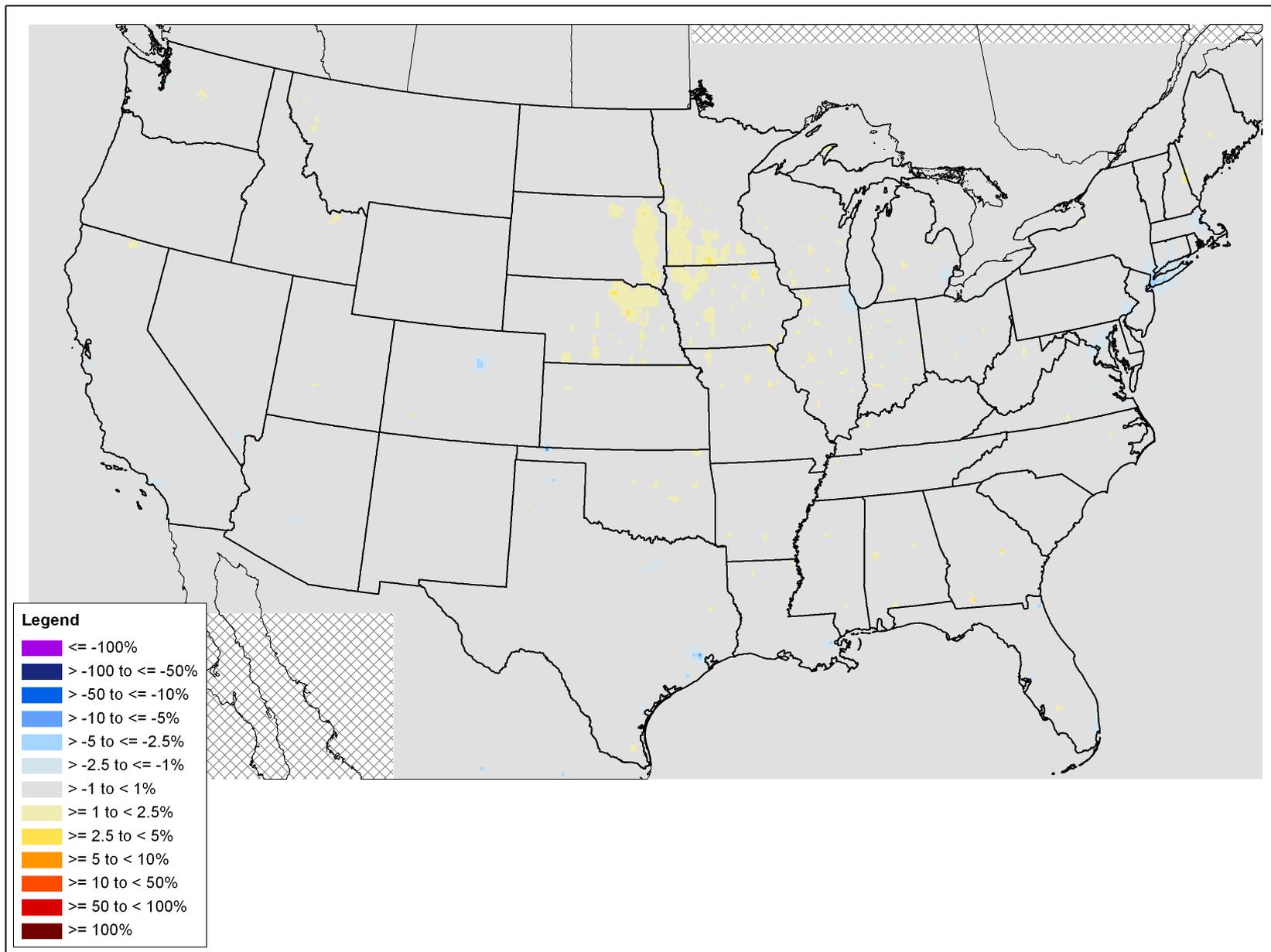
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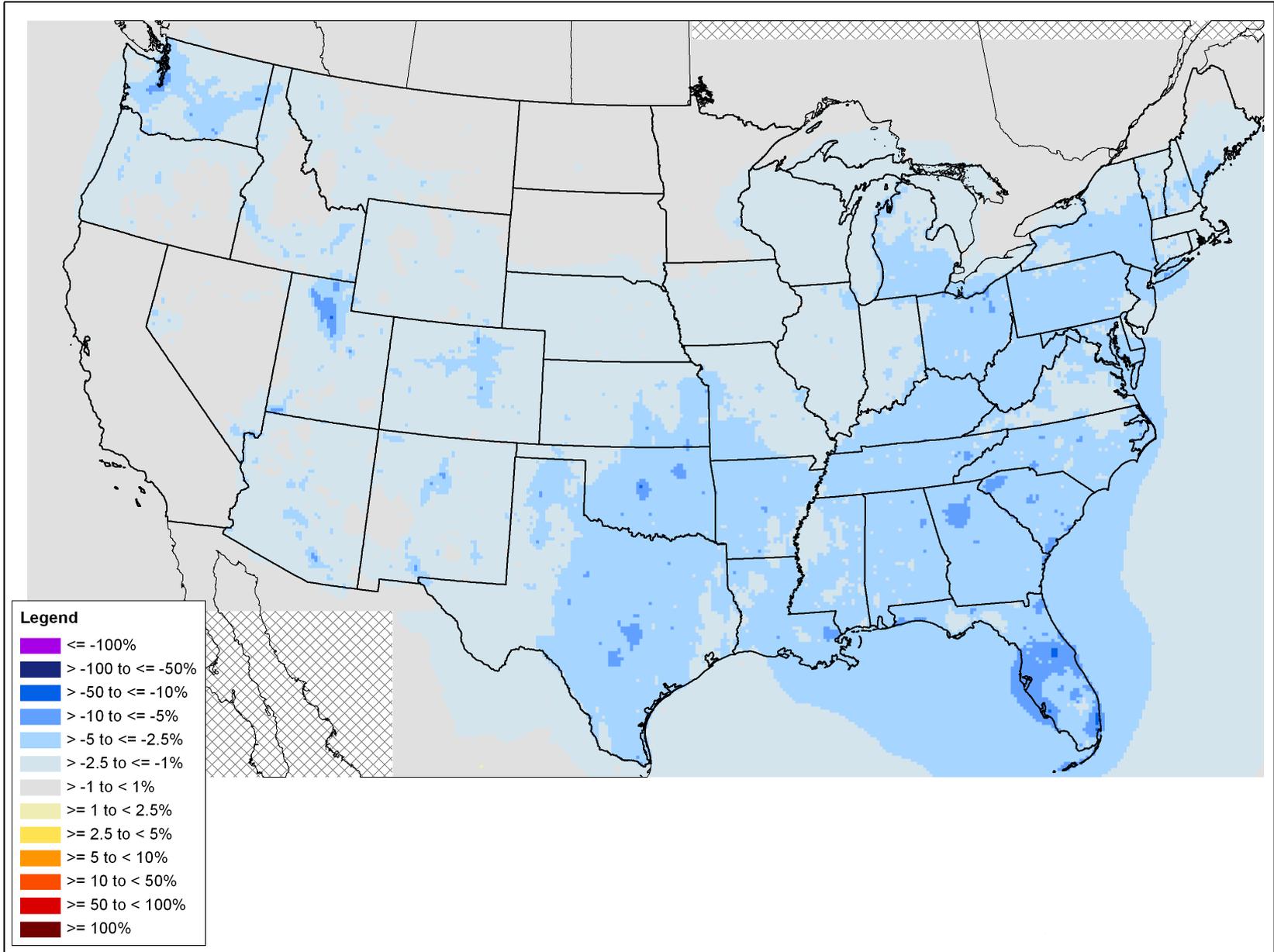
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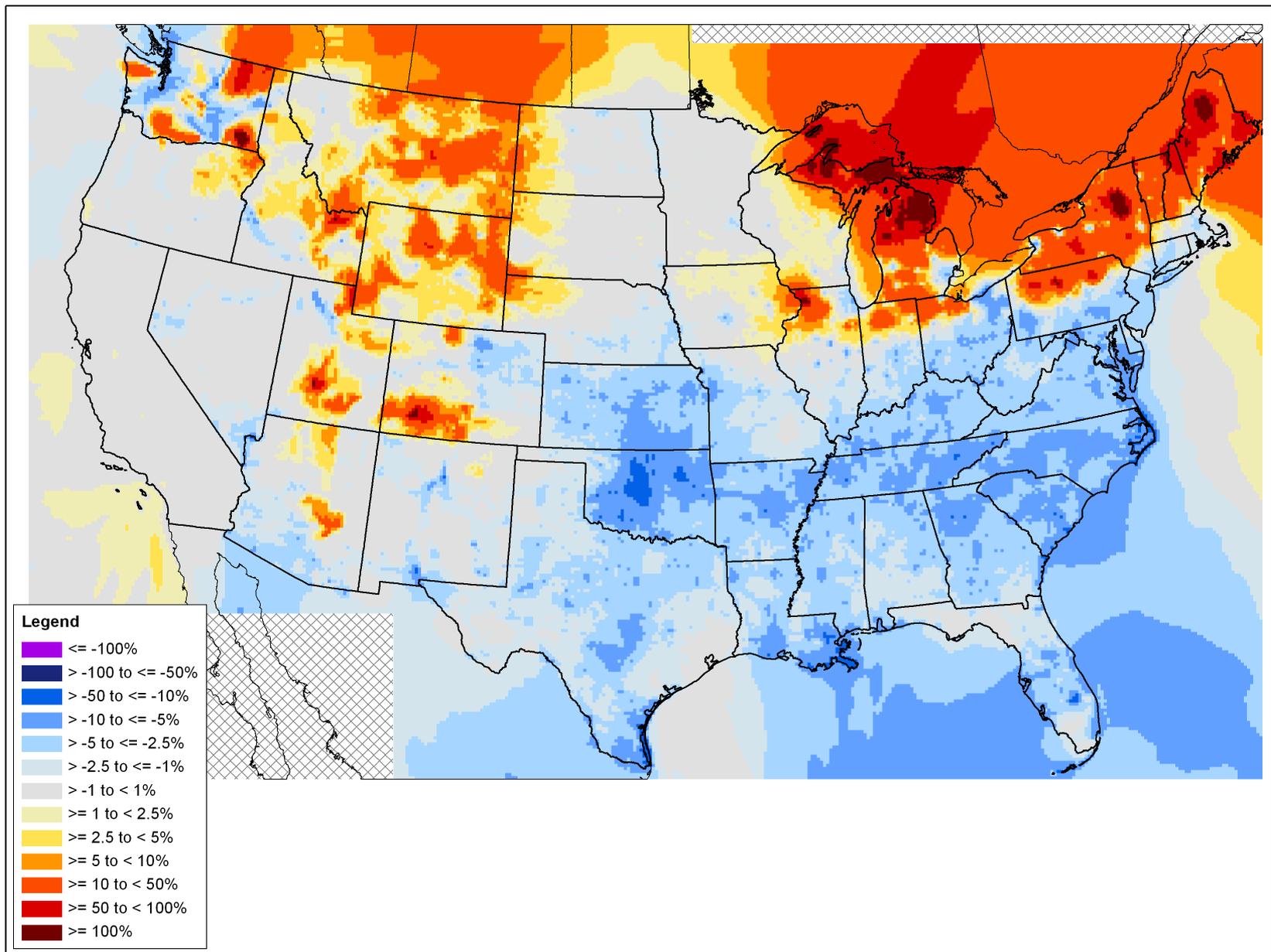
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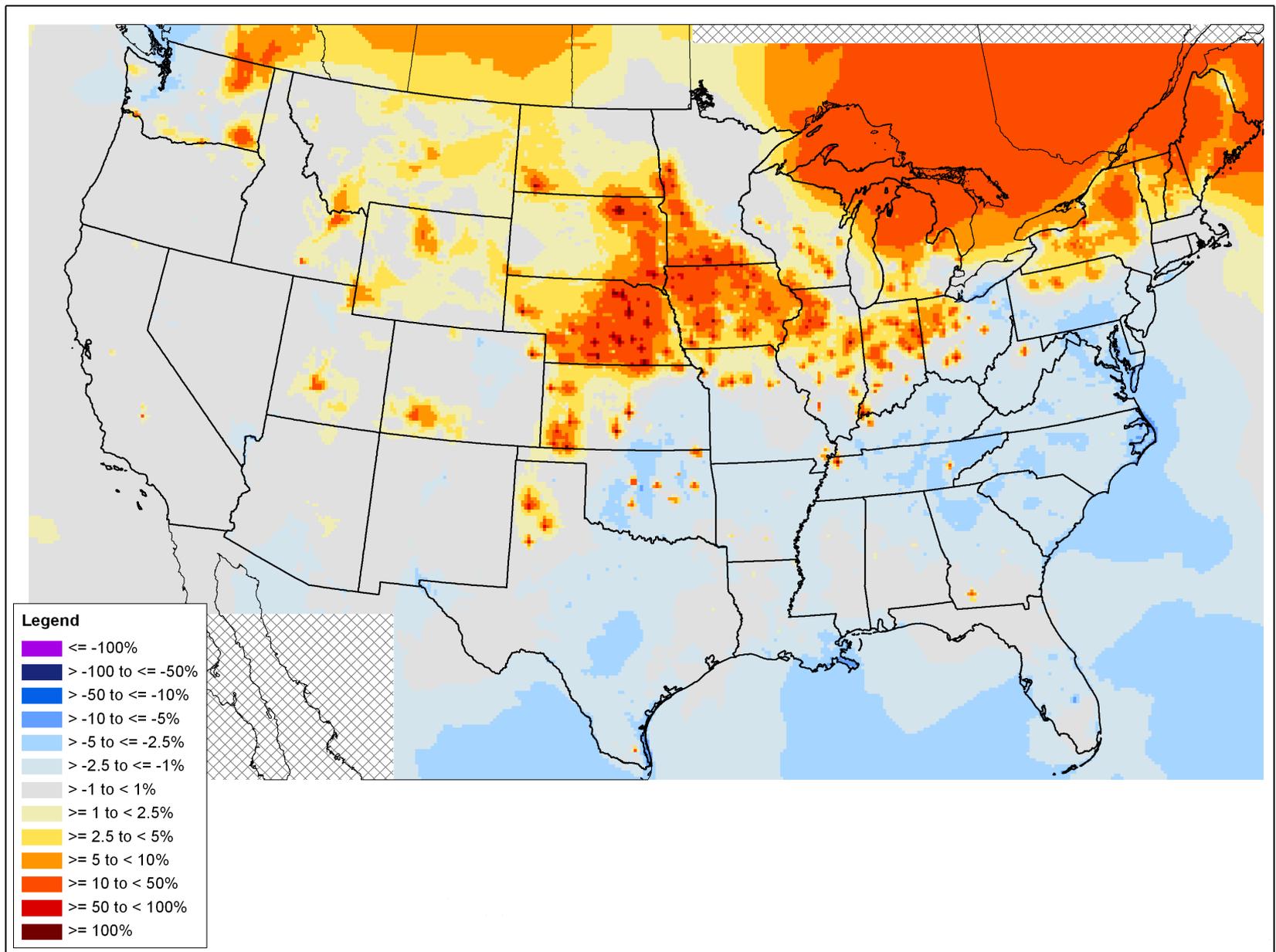
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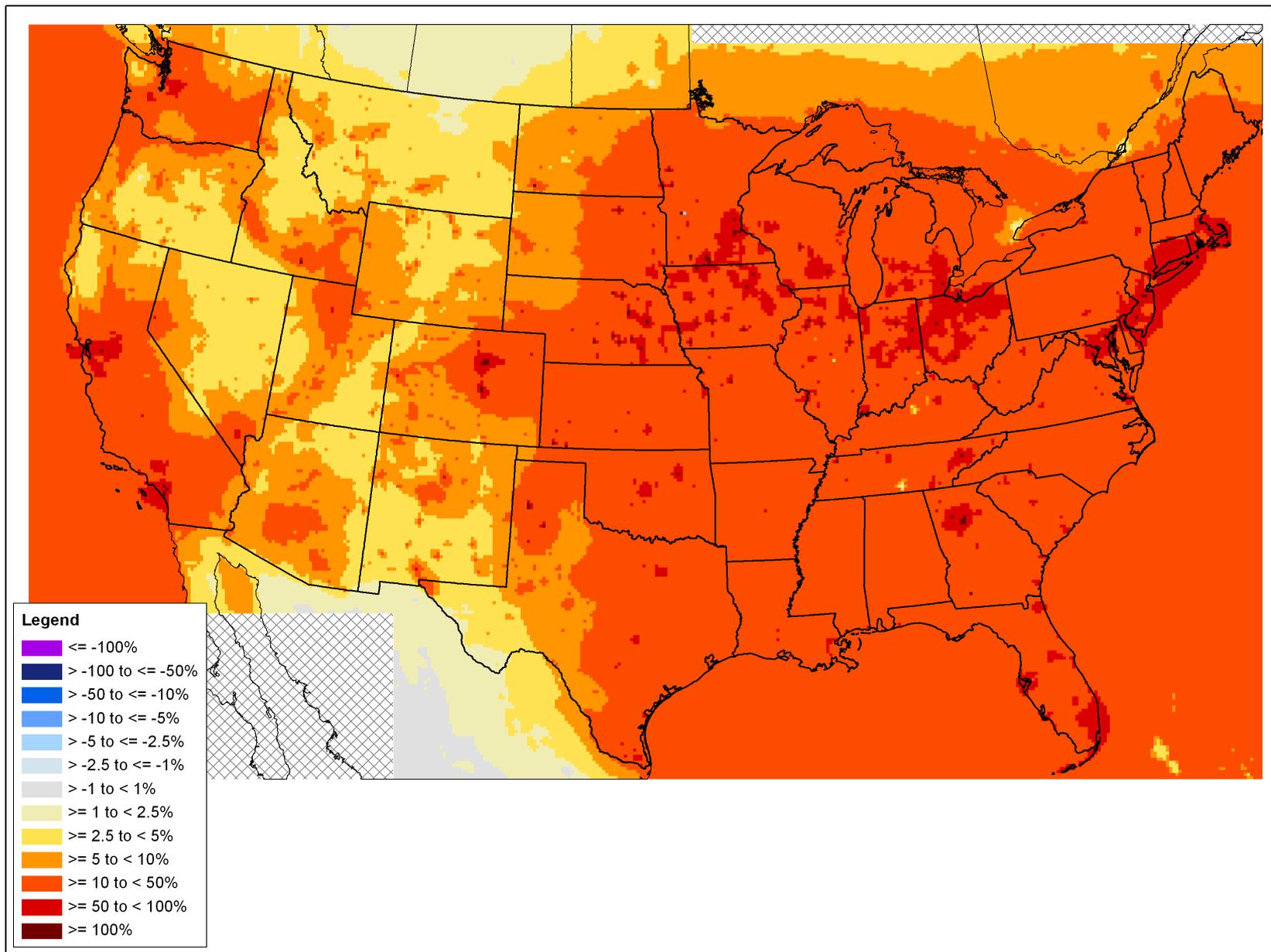
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