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**Comments on Hill et al. "Air Quality Related Health Damages of Maize"**

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Introduction

Hill et al. in "Air Quality Related Health Damages of Maize" published April 1, 2019 in *Nature Sustainability* report that "maize production is associated with 4,300 premature deaths annually" in the United States. In that paper these deaths are attributed to fine particulate matter (PM2.5) of primary and secondary origin emitted on a life cycle basis along the production pathway of corn. Primary PM2.5 emissions are from fuel combustion, dust with particle size less than 2.5 micrometers whereas secondary emissions refer to the conversion under certain geographic and climatic conditions of other pollutants (including VOCs, $NH_3$, $NO_x$, and $SO_x$) into additional PM2.5 particles. The paper attributes most of the mortalities to secondary emissions and states that "increased concentrations of fine particulate matter (PM2.5) are driven by emissions of ammonia—a PM2.5 precursor—that result from nitrogen fertilizer use."

In life cycle modeling researchers distinguish between tank to wheel (TTW) emissions which include vehicle evaporative emissions and vehicle tailpipe combustion emissions and well-to-tank (WTT) emissions, which include fuel production and distribution. The TTW and WTT emissions combined result in well-to-wheels emissions (WTW). The most commonly used lifecycle model in the US is the Greenhouse gases, Regulated Emissions, and Energy use in Transportation Model (GREET) developed by Argonne National Laboratory. That model includes both WTT and TTW emissions. Methodologically, Hill et. al. re-parameterize a portion of the WTT section of the GREET model (the section related to corn agriculture) and combine its output with an atmospheric model that is unrelated to GREET to derive life cycle particulate matter emissions for corn production and life cycle health impacts in the form of mortalities. Greenhouse gases and economic cost are tracked as an additional metric.

The Hill paper provides some potentially important insights into the contribution of secondary emissions to human health effects. But we question the appropriate placement of this work within life cycle methodology. ISO 14044 provides guidance on life cycle modeling as follows: "Systems shall be compared using the same functional unit and equivalent methodological considerations, such as performance, system boundary, data quality, allocation procedures, decisions rules…."[1] Hill et al. state:

> "Our focus here has been on the production of maize, but nearly all maize is transformed before its final use by consumers. Nearly 90% of maize grown in the United States is used for animal feed or ethanol biofuel, both of which lead to further emissions of primary PM2.5 and secondary PM2.5 precursors, as well as GHGs […]"

Since Hill et al. directly say that the main products of corn are feed and biofuels then the correct life cycle boundaries would be around those end products. Looking at just individual life cycle stages introduces serious comparative data quality issues as we will show later.

[1] ISO 14044 Environmental Management – Life Cycle Analysis

Hill et al. point out corn is indeed produced to provide feed, food, and biofuels. If corn is not grown then alternative feed, food, or fuel products would be grown which would also emit pollutants and cause mortalities. This means we should quantify the emissions and mortalities of products for which corn substitutes. A large amount of corn acres (35%) produces both corn ethanol as well as coproduct feed.[2] In life cycle analysis then we need to assess both the displaced emissions and mortalities (whether greenhouse gas emissions or primary or secondary air pollutant emissions) in the feed markets as well as the biofuel ethanol markets. And, as laid out in the ISO guidelines, the comparison has to made with the same data quality requirements for corn products as well as its substitutes. One useful life cycle framework, for example, would have been the direct comparison between corn ethanol production (including obviously corn production as one life cycle stage) and gasoline. This would have allowed to control for consistent data quality along both pathways.

Comparative Example of Life Cycle Modeling BasedGHG Emissions

Figure 1 below shows total life cycle GHG emissions for corn ethanol production which enables a direct comparison with total life cycle GHG emissions for gasoline production. The units are carbon dioxide equivalent emissions per heating unit ($gCO_2e/MJ$ or sometimes $gCO_2e/MMBTU$). The figure also shows the relative emissions contributions (in percent) from each life cycle stage to the total. The total life cycle emissions for corn ethanol of 62 $gCO_2e/MJ$ contain a significant coproduct credit of 14 $gCO_2e/MJ$ for the agricultural phase since only the starch in the corn kernel is converted to ethanol with the rest going into the feed and food markets in the form of distillers grains and solubles (DGS). GHG emissions from land use change are included in the assessment. As a finished product then the 62 $gCO_2e/MJ$ emitting ethanol on a WTW basis as a fuel displaces life cycle GHG emissions of gasoline which range from 90-100 $gCO_2e/MJ$. Note that for the corn ethanol GHG life cycle the TTW emissions are a very small contribution since most emissions are incurred during the production process and the carbon in the fuel is carbon neutral, since the carbon is from the air during corn growth.

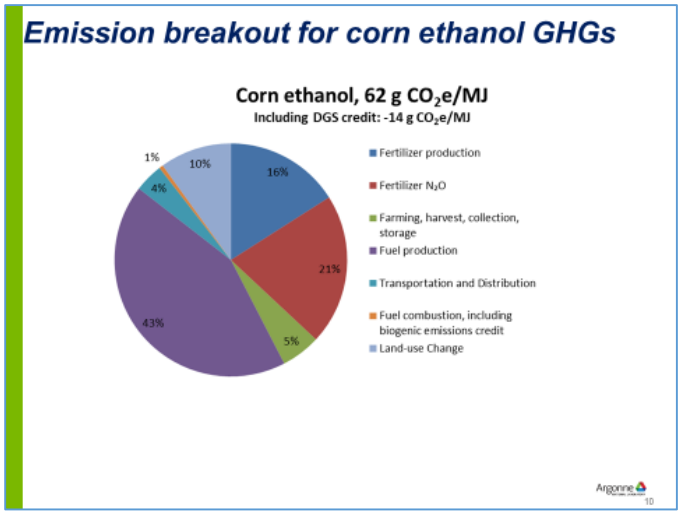


Figure 1: Corn ethanol GHG Life Cycle Results (Source: M. Wang. Argonne Presentation May 2019)

[2] http://www.worldofcorn.com/#corn-usage-by-segment

Life Cycle Emissions of Other Key Pollutants

For primary pollutants other than GHGs the table below shows the current GREET WTT emissions for ethanol compared to gasoline. Note that these emissions (as with GHG emissions) are again presented on a mass-of-pollutant-per-heating-unit basis (g/MMBtu) for all life cycle stages including for ethanol the emissions associated with corn production and corn conversion at the ethanol plant with proper coproduct credit given to DGS coproducts. For the gasoline pathway emissions incurred during petroleum recovery and refining, gasoline transportation and distribution, and others are included. Note that the table does not include emissions from combustion in the vehicle (discussed later) so these partial life cycle stages are referred to as WTP emissions. As can be seen in the table, the WTT emissions (highlighted columns) which include the corn production phase are substantially lower for all assessed pollutants for ethanol compared to gasoline. This demonstrates that potential misleading conclusion if one only considers a stage, not the entire life cycle of a final product and if one does not conduct a comparative analysis.

Table 1: Life cycle pollutant emissions for corn ethanol and gasoline by life cycle stage (Source: Life Cycle Associates)

| g/MMBtu | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | Ethanol | | | Gasoline | | | | | | | |
| | Corn | Ethanol | WTT | Gasoline Blendstock Refining: Feed Inputs | Gasoline Blendstock Refinig: Intermediate Product Combustion | Gasoline Blendstock Refining: Non-Combustion Emissions | Gasoline Blendstock Transportation | Gasoline Blendstock Distribution | Gasoline Distribution | Gasoline Storage | WTT |
| Urban VOC | 0.168 | 13.567 | 13.736 | 0.374 | 0.221 | 1.762 | 13.242 | 0.021 | 0.022 | 0.000 | 15.643 |
| Urban CO | 0.287 | 1.631 | 1.919 | 0.550 | 1.531 | 0.881 | 0.049 | 0.065 | 0.068 | 0.000 | 3.144 |
| Urban NOx | 0.767 | 4.013 | 4.780 | 1.013 | 2.609 | 1.042 | 0.225 | 0.158 | 0.165 | 0.000 | 5.211 |
| Urban PM10 | 0.072 | 0.424 | 0.496 | 0.148 | 0.566 | 0.244 | 0.018 | 0.006 | 0.006 | 0.000 | 0.988 |
| Urban PM2.5 | 0.058 | 0.341 | 0.400 | 0.120 | 0.527 | 0.122 | 0.015 | 0.003 | 0.003 | 0.000 | 0.790 |
| Urban SOx | 0.588 | 4.534 | 5.121 | 1.170 | 0.734 | 3.369 | 0.141 | 0.006 | 0.006 | 0.000 | 5.425 |
| Urban BC | 0.006 | 0.019 | 0.025 | 0.011 | 0.075 | 0.000 | 0.002 | 0.000 | 0.000 | 0.000 | 0.088 |
| Urban OC | 0.014 | 0.056 | 0.070 | 0.017 | 0.091 | 0.000 | 0.004 | 0.000 | 0.000 | 0.000 | 0.112 |

These pollutants are also tracked by Hill et al. for the corn phase as shown in the table and then converted to mortalities for PM2.5 with one important distinction: As mentioned above, Hill et al. attribute significant emissions and mortalities to ammonia from fertilizer which converts to particulate matter (PM2.5) in the atmosphere as a secondary pollutant. Hill et al stated “The dominant contributor to maize production-related PM2.5 concentrations is emissions of NH3 from synthetic nitrogen fertilizer and manure application […], which account for 71% of attributable deaths.” The atmospheric modeling component will be described in the next section.

Atmospheric Modeling of Secondary Emissions for Corn/Ethanol Life Cycle Emissions

As pointed out above, Hill et al. derive the PM2.5 emissions (in mass) from maize production using GREET and convert this into concentrations using a separate atmospheric model. In health-effect studies, the mass of air emissions pollutants is indeed generally converted into atmospheric concentrations (selected models exist that take geography, wind speed, source proximity and other

parameters into consideration) to allow a comparison for each considered pollutant against concentration-health response functions (many studies establish these relationships based on animal or cohort studies).

The science behind precursor PM emissions (the focus of Hill et al.) in atmospheric modeling points to significant uncertainties. As stated in a report prepared for the Federal Highway Administration (p. 3)[3]: "there is general consensus that the formation of PM2.5 from precursor compounds is highly uncertain and varies regionally and seasonally due to weather conditions and other related variables that affect atmospheric chemistry." With respect to emissions components that are field-related the FHA report states (page 1) "the portion of PM2.5 comprised of all secondary components (sulfates, nitrates, ammonium, organic carbon) varies anywhere from 30% to 90% of all PM2.5."

In their work, Hill et al. state that they are using the InMap Model for atmospheric modeling.

> "We employed the Intervention Model for Air Pollution (InMAP) to conduct reactive dispersion air quality modelling and to estimate the premature mortality in the United States attributable to maize production. InMAP creates spatially explicit estimates of the effect of emissions of primary PM2.5 and secondary PM2.5 precursors (VOCs, NOx, NH3 and SOx) on atmospheric concentrations of PM2.5 [...]"

But InMap's own publication seems to refer to the uncertainties related to secondary PM2.5 emissions and states:[4]

> "For $NO_x$, $NH_3$, and VOCs, the chemical reaction mechanisms governing partitioning between the gas and particle phase are more complex than the reactions driving sulfate formation. They are also reversible: gas-phase compounds can convert to aerosols and then back to gas-phase, and the direction of the reactions can vary according to the time of day and according to the season. It is not possible to directly represent these reactions in a steady-state, annual average model such as InMAP."

Importantly, InMap was also developed by Hill as one of the lead authors.[5] It is unclear to what extent the uncertainties related to PM2.5 precursor attributions pointed out in the Federal Highway document are considered in this model. InMap is not listed on the EPA website under "Air Quality Dispersion Modeling - Preferred and Recommended Models."[6]

[3] William Hodan and Willliam Barnard. "Evaluating the Contribution of M2.5 Precursor Gases and Re-entrained Road Emissions to Mobile Source PM2.5 Particulate Matter Emissions". https://www3.epa.gov/ttnchie1/conference/ei13/mobile/hodan.pdf

[4] "InMAP: A model for air pollution interventions"; Christopher W. Tessum, Jason D. Hill, Julian D. Marshall PLoS One. 2017; 12(4): e0176131. Published online 2017 Apr 19. https://www.ncbi.nlm.nih.gov/pmc/articles/PMC5397056/#pone.0176131.s017

[5] http://spatialmodel.com/inmap/#home

[6] Air Quality Dispersion Modeling - Preferred and Recommended Models

In other words, Hill's mortality number and the key contributions from the paper rely on highly uncertain science and an internally developed atmospheric model. The uncertainty, however, is never mentioned in the paper.

Atmospheric Modeling of Secondary Emissions for Gasoline Life Cycle Emissions

In this section we point out that it is important to employ consistent data quality in life cycle analysis and that only correct modeling boundaries enable comparative assessments between technology pathways. As with secondary emissions during corn production, secondary emissions are also a topic of evolving science downstream in the life cycle of the comparative corn ethanol/gasoline pathway (PTW portion). A paper coauthored with US EPA describes that secondary organic aerosols (SOAs) are a major contributor to PM2.5 with aromatics in gasoline being in turn the most effective precursors to SOAs.[7]

> "Field studies suggest 10% - 60% of fine particulate matter (PM2.5) is comprised of organic compounds. This material may be directly emitted to the atmosphere (primary) or formed from the gas-phase oxidation of hydrocarbon molecules and subsequent absorption into the condensed phase (secondary). The latter portion, referred to as secondary organic aerosol (SOA), is a major contributor to the PM2.5. Evidence is growing that aromatics in gasoline exhaust are among the most efficient secondary organic matter precursors. While the relative abundance of primary and secondary organic matter is the subject of ongoing debate, air quality models are continually updated to keep up with the latest scientific knowledge […]. In the United States, gasoline-powered vehicles are the largest source of aromatic hydrocarbons to the atmosphere. Most gasoline formulations consist of approximately 20% aromatic hydrocarbons, which are used in place of lead to boost octane. Therefore, it has been suggested that removal of aromatics could reduce SOA concentrations and yield a substantial public health benefit."

Figure 2 in that EPA-coauthored paper presents a nationwide map of predicted premature mortalities attributable to aromatic hydrocarbons in gasoline associated with the expert elicitation concentration-response function. Predicted premature mortalities range from nearly 1,850 to more than 4,700 cases, depending on which concentration-response function is used, and to over 5,000 up to 6,300 cases when adjusted for urban shares.

The evolving science in this area is important because the EPA Fuels Trends Report (released in November 2017) shows reduction in aromatics since 2000 of 24% and directly attributes the reductions to ethanol.[8] On page 8 that report states: "Ethanol's high octane value has also allowed refiners to significantly reduce the aromatic content of the gasoline, a trend borne out in the data."

Moreover, setting the life cycle modeling boundaries around ethanol compared to gasoline would have also allowed Hill et al. to include some of the latest research on direct PM emissions all the way upstream. That research points to an overall reduction of direct PM emissions from ethanol blended

[7] Public health impacts of secondary particulate formation from aromatic hydrocarbons in gasoline; Katherine von Stackelberg, Jonathan Buonocore, Prakash V Bhave & Joel A Schwartz Environmental Health Volume 12, Article number: 19 (2013)

[8] Fuel Trends Report: Gasoline 2006 - 2016 ; Office of Transportation and Air Quality; U.S. Environmental Protection Agency; EPA-420-R-17-005; October 2017; https://nepis.epa.gov/Exe/ZyPDF.cgi?Dockey=P100T5J6.pdf

gasoline particularly for the dominant gasoline direct injection engine type which is the dominant combustion engine type going forward.[9;10;11;12;13]

Summary

The Hill et al. paper intends to emphasize indirect PM emissions from corn agriculture in life cycle analysis. But by setting incomplete life cycle boundaries, the paper does not allow a direct comparison with indirect emission (and direct emissions) from comparative products which could be much worse alternatives. The main comparative product, gasoline, incurs substantial indirect PM emissions post-combustion in the atmosphere and higher direct emissions during combustion. So the emissions benefits of the key substituting corn product (ethanol) were not considered. Furthermore, the paper relies heavily on an internally developed atmospheric model (as opposed to EPA preferred and recommended models) with documented high uncertainties in modeling these type of emissions. Finally, we find that the scarce material in the supporting information made it very difficult to attempt a verification of the GREET parameterization (e.g. employed fertilizer usage rates) of the employed atmospheric model (concentration modeling assumptions etc.).

[9] Munoz et al. “Bioethanol Blending Reduces Nanoparticle, PAH, and Alkyl- and Nitro-PAH Emissions and the Genotoxic Potential of Exhaust from a Gasoline Direct Injection Flex-Fuel Vehicle”; Environ Sci Technol. 2016 Nov 1;50(21):11853-11861. 2016 Oct 20. Swiss Federal Laboratories for Materials Science and Technology

[10] K. Aikawa and J. J. Jetter, "Impact of gasoline composition on particulate matter emissions from a direct-injection gasoline engine: Applicability of the particulate matter index," International Journal of Engine Research, vol. 15, no. 3, pp. 298-306, 24 June 2013.

[11] Jin, D., Choi, K., Myung, C.L., Lim, Y., Lee, Y., Park, S., 2017. The impact of various ethanol-gasoline blends on particulates and unregulated gaseous emissions characteristics from a spark ignition direct injection (SIDI) passenger vehicle. Fuel. http:// dx.doi.org/10.1016/j.fuel.2017.08.063.

[12] Storey, J. M., Barone, T., Norman, K., and Lewis, S. 2010. Ethanol Blend Effects on Direct Injection Spark-Ignition Gasoline Vehicle Particulate Matter Emissions. SAE Technical Paper No. 2010-01-2129. SAE, Warrendale, PA.

[13] Martini, G., Astorga, C., Adam, T., Farfaletti, A., Manfredi, U., Montero, L., Krasenbrink, A., Larsen, B. and De Santi, G. Effect of Fuel Ethanol Content on Exhaust Emissions of a Flexible Fuel Vehicle, JRC Report 2009