Climate impacts of short-lived climate forcers versus CO_2 from biodiesel: A case of the EU on-road sector

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Abstract

Biofuels are proposed to play an important role in several mitigation strategies to meet future CO_2 *emission targets for the transport sector, but remain controversial due to significant uncertainties in net impacts on environment, society and climate. A switch to biofuels can also affect short-lived climate forcers (SLCFs), which provide significant contributions to the net climate impact of transportation. We quantify the radiative forcing (RF) and global-mean temperature response over time to EU on-road fossil diesel SLCFs, and the impact of 20% (B20) and 100% (B100) replacement of fossil diesel by biodiesel. SLCFs are compared to impacts of on-road* CO_2 *using different approaches from existing literature to account for biodiesel* CO_2*. Given the best estimates for changes in SLCFs when replacing fossil diesel with biodiesel, the net positive RF from EU on-road fossil diesel SLCF of 3.4* mW/m^2 *is reduced by 15% and 80% in B20 and B100, respectively. Over time the warming of SLCFs is likely small compared to biodiesel* CO_2 *impacts. However, SLCFs may be relatively more important for the total warming than in the fossil fuel case if biodiesel from feedstock with very short rotation periods and low land-use-change impacts replaces a high fraction of fossil diesel.*

Introduction

Multiple alternative vehicle and fuel options to reduce the emissions and climate impact of the transport sector have been proposed. This study explores one such option – replacement of conventional fossil diesel with biodiesel. Biofuels (referring to liquid or gaseous fuels derived from biomass) currently provide around 2% of the global transport fuel, with higher shares in certain countries.[1] However, biofuels are proposed to play an important role in several mitigation strategies for meeting future emission targets for the transport sector. For instance, the European Union (EU) Renewable Energy Directive (RED) includes a 10 percent target for renewable energy in transportation in every member state by 2020.[2] Similarly, the U.S. Renewable Fuel Standard (RFS2) program under the 2007 Energy Independence and Security Act requires 36 billion gallons, about 7% of expected annual gasoline and diesel consumption, of renewable fuel to be blended into transportation fuel by 2022.[3] The International Energy Agency estimate that biofuels could provide 27% of the global transport fuel by 2050,[1] while the Nordic Energy Outlook project biofuel shares of total fuel from 25% to 70% by 2050 depending on scenario.[4] Despite their significant role in mitigation strategies, biofuels remain controversial because the net impact on the environment, society and climate can be difficult to determine.[5]

The role of biofuels in reducing greenhouse gas (GHG) emissions is generally evaluated using the life-cycle assessment (LCA) methodology. The standard practice in LCA of climate impact is to compare emissions of long-lived GHGs using the Global Warming Potential (GWP) metric with a time horizon of 100 years, consistent with the Kyoto Protocol framework.[6] Traditionally biofuels were considered carbon, and hence climate, neutral over the life cycle because of the assumption that CO_2 released from combustion approximately equals the CO_2 sequestered in the biomass. However, a number of studies have shown that emissions from direct and indirect land-use change (LUC) can make carbon footprints of biofuels highly positive, i.e., biofuels have a warming climate impact.[7-10] The carbon and climate neutrality assumption also ignores important factors such as the temporary climate impact of biogenic carbon between the time of its release to the atmosphere by biofuel combustion and its sequestration during feedstock regrowth,[11,12] as well as changes in surface albedo.[13,14] Hence, the role of biofuels in reducing the GHG emissions from the transport sector – and the consequent climate impact – is determined by a number of factors, and several different approaches to account for biomass CO_2 and LUC impacts exist in the literature.

In addition to CO_2, the transport sector is an important source of short-lived climate forcers (SLCFs: in this study comprising aerosols, ozone and methane). These make important warming and cooling contributions the total climate impact and act on very different temporal scales.[15] Aside from CO_2, the main contributions to *warming* from road transport are from emissions of black carbon aerosols (BC) and from ozone (O_3) produced by emissions of carbon monoxide (CO), nitrogen oxide (NO_x) and volatile organic carbon (VOCs). *Cooling* impacts are caused by NO_x-induced reductions in methane (CH_4) and by organic carbon (OC), sulfate (SO_4) and nitrate aerosols.

Replacing conventional fossil fuels with biofuels affect the tailpipe emissions of gases and aerosols and the impact of diesel-biodiesel blends on the exhaust emissions of regulated species has been extensively studied.[16-18] Biofuels are essentially sulfur free and hence reduce emissions of SO_2. The majority of studies show clear reductions in tailpipe emissions of CO, hydrocarbons and particulate matter (PM) with biodiesel use. The effect on NO_x emissions is more difficult to assess, although the average of available studies point to a slight increase with biodiesel. Considerably less attention has been given to the climate impact of SLCFs following such emission changes, although two recent studies have looked at the climate impact of biofuels in the aviation and shipping sectors.[19,20]

In this study we apply a global chemistry-transport model to quantify the global-mean radiative forcing (RF) of SLCFs due to emissions from the on-road fossil diesel sector and the impact of replacing conventional fossil diesel by biodiesel. We select the EU as a case, mainly due to the high share of diesel in the total fuel consumption and specific target for renewable fuels by 2020. Furthermore, the global-mean temperature response over time to the RFs is calculated. Sustained replacement of fossil diesel with biodiesel will affect both SLCFs and long-lived GHGs, and it is crucial to place the impacts on a common scale to facilitate a proper comparison of the impacts. By using time dependent global-mean temperature response we illustrate the relative importance of CO_2 and SLCFs over time. The temperature response to biodiesel CO_2 is estimated under several different assumptions about how to account for CO_2 from biomass sources, with the aim of assessing under which conditions SLCFs might be important compared to CO_2 and reflecting the significant uncertainty in the existing literature. The impact of assuming different feedstock rotation periods, LUC emissions and biofuel blends is explored.

Methodology

This section gives a brief description of the methodology. Further details and a flowchart are provided in the Supporting Information (SI – Sections S1 and S2 and Figure S1).

Emissions and chemistry-transport modeling

To simulate the contribution to atmospheric concentrations of aerosols and gases resulting from emissions from the current (i.e., year 2010) EU on-road fossil diesel sector ("FF") and the changes in concentrations when fossil diesel is replaced by biodiesel, the chemistry transport model OsloCTM2 with a microphysical aerosol parameterization is used.[21,22] The emissions of on-road fossil diesel CO, VOCs, NO_x, SO_2, ammonia (NH_3), BC and OC have been developed with the GAINS model (http://gains.iiasa.ac.at) as part of the ECLIPSE project funded by the European Commission 7th Framework Programme.[23-25]

To investigate the impact of a replacing fossil diesel with biodiesel, two idealized biofuel cases are defined. The first case assumes that the entire EU on-road diesel sector has a 20% (by energy) biodiesel blend with fossil diesel ("B20") and the second assumes a 100% replacement of fossil diesel by biodiesel ("B100"). The total fuel consumption is kept constant at the 2010 level and instantaneous replacement of fossil diesel is assumed. Changes in on-road emissions of aerosols, CO, NO_x and VOCs when fossil diesel is replaced by biodiesel are taken from the review by the US Environmental Protection Agency (EPA).[26] Using a large amount of emissions data from the 1980s and 1990s, the EPA derived relationships expressing the best fit between percentage changes in exhaust emissions and biofuel blend. The emissions data was limited to North American engines and primarily heavy-duty vehicles. Moreover, emission factors for new vehicles have changed significantly during recent years. However, a more recent review show that the relationships generally hold also when measurements from newer European and Japanese light-duty vehicles are included, although a somewhat lower reduction of CO and hydrocarbon emissions is seen on average for high blends.[16] The impact of biodiesel on exhaust emissions depends on e.g., vehicle/engine characteristics, driving conditions and biomass feedstock. It is important to note that the relationships express the best fit; there is a considerable range in measurements for all blends.

Table 1 summarizes year 2010 EU on-road fossil diesel emissions and the percentage emission changes from a switch to biodiesel assumed in our cases. For each species and case, the total emission is scaled, keeping the spatial distribution constant. Ammonia (NH_3) is accounted for in the OsloCTM2 and fossil diesel NH_3 emission, albeit small, are included in the emission inventory. However, due to lack of information we do not account for changes in NH_3 emissions with biodiesel blends.

TABLE 1

Climate impact calculations

The global-mean radiative forcing (RF) of aerosols (BC, OC, SO_4 and nitrate) is estimated using the 3-dimensional changes in concentrations from the OsloCTM2 with normalized forcing distributions from Samset and Myhre.[27] Forcing from indirect aerosol effects and the deposition of BC on snow is not included. The RF of the NOx/CO/VOC-induced change in O_3 concentrations is calculated using a 2-dimensional normalized forcing distribution.[28] Emissions of NO_x, CO and VOCs also affect the lifetime and concentration of CH_4, which gives a consequent perturbation in O_3.[29] The RF of NO_x/CO/VOC-induced changes in CH_4 is calculated from the global-mean change in methane lifetime as described in Section S1 and the RF of the subsequent methane-induced O_3 loss is calculated as $0.5 \cdot RF_{CH4}$.[30]

The global-mean temperature response over time to the RFs of fossil diesel and biodiesel SLCFs from the EU on-road diesel sector is calculated using the impulse response function (IRF_T) from Boucher and Reddy.[31] The temporal evolution of SLCFs is assumed to follow a simple exponential decay with one time scale.[32,33] To calculate the temporal evolution of the atmospheric fossil CO_2 concentration, the impulse response function (IRF_{CO2}) from Joos et al.[34] is used. The resulting normalized temperature response is multiplied by CO_2 emissions to estimate the impact of the EU on-road sector. On-road emissions of CO_2 are calculated from total diesel consumption in the GAINS model (8000 PJ in 2010; Zbigniew Klimont, personal communication) using a specific CO_2 emission factor of 73.2 g/MJ for fossil diesel.[35] Calculations of temperature impacts of biodiesel CO_2 are described below. Two different temporal perspectives wrt emissions are illustrated. First, we consider the temperature response over time to pulse (i.e., one year) emissions from the EU on-road diesel sector. Pulses are useful for illustrating the different temporal behavior of various mechanisms contributing to the temperature response of a sector. Moreover, pulses can also be used by convolution to construct any other kind of scenario.[15] Using this approach we also calculate the temperature response to sustained constant year 2010 emissions, which illustrates continuous climate impacts from emissions in an idealized no-growth scenario.

Accounting for biodiesel CO_2

To put the estimated temperature responses to SLCFs in context, we compare these with some simplified estimates of the response to biodiesel CO_2. To calculate the temperature response to biodiesel CO_2 from the EU on-road sector we define five illustrative cases using different approaches based on existing literature on how to account for CO_2 from biomass sources (see Section S2 for additional details), which are summarized in Table 2. First, we adopt the assumption that biodiesel is carbon neutral, i.e., no net CO_2 emissions. This case is included purely for illustrative purposes. As noted above, a number of studies have disproved the carbon-neutrality assumption. Nevertheless, a number of LCA studies and guidance for carbon footprinting have presumed that biomass is carbon neutral (e.g., Johnson[36]) and it is useful to illustrate the effect of this assumption. Next we illustrate potential net CO_2 emission savings, i.e., when including LUC emissions, from biodiesel relative to fossil diesel and the following temperature response. In these cases, two symmetric net savings factors, selected to illustrate the effect of high and low LUC impacts, are used to calculate CO_2 emissions from the EU on-road diesel sector. Finally we replace the IRF_{CO2} with the IRF for biogenic carbon (IRF_{bio}) from Cherubini et al.[12] in the calculation of the temporal evolution of atmospheric CO_2 from biodiesel. This function accounts for the time lag between the release of biomass carbon by combustion and its uptake during biomass regrowth, determined by the feedstock rotation period, during which time the CO_2 released to the atmosphere will have a climate impact.

TABLE 2

Results

This section presents first the changes in atmospheric concentrations and RF of SLCFs resulting from the changes in emissions due to a switch to biodiesel. Next the global-mean time dependent temperature responses to SLCFs from the current EU on-road fossil diesel sector, and from the sector after the replacements of fossil diesel by biodiesel, are presented. Finally, the temperature response to SLCFs is compared with the response to CO_2.

Biodiesel impacts on concentrations and RF of SLCFs

Figure 1 shows the modeled changes in annual mean atmospheric burden of BC, SO_4, nitrate aerosol and O_3 resulting from the changes in emissions when fossil diesel in the EU on-road sector is 100% replaced by biodiesel (B100). For comparison, modeled burdens resulting from the current EU on-road fossil diesel emissions are showed in Figure S2.

Emissions from the current EU on-road fossil diesel sector results in a BC burden of up to 150 μg/m^2 over central Europe (Fig. S2a), which constitutes 15-35% of the total modeled BC burden over much of the region. Replacing fossil diesel with biodiesel reduces the BC burden (Fig. 1a) and reductions up to 80 μg/m^2 can be seen in the B100 case. The reductions are found to scale quite linearly with the emission reduction. The on-road diesel sector is a much smaller source of OC than BC, providing only up to 6% of the total OC burden over central Europe (not shown here). Similarly to BC, the burden of OC is reduced by up to 50% in B100 compared to the fossil fuel case. On-road fossil diesel emissions cause both increases and decreases in the annual average burden of SO_4, with the strongest increase seen over the Mediterranean and the decrease mainly localized to western Europe (Fig. S2b). This is a result of emissions of both SO_2 and ozone precursors, as well as local background meterological conditions, and there can be significant variability in the sign of the SO_4 response to ozone precursor emissions among different models, as illustrated by Fry et al.[37] for emissions from all sources. Replacing fossil diesel with biodiesel gives a small decrease in the burden of SO_4 (Fig. 1b). Moreover, because the effect of biodiesel on SO_4 is determined by changes both in SO_2 emissions and the atmospheric oxidation capacity, the burden change does not scale linearly with the strong SO_2 emission reduction of 90% assumed in the B100 case. Biodiesel has low sulfur content, hence reducing the SO_2 available for production of SO_4. Simultaneously, the reductions in emissions of CO and VOC and the increase in NO_x emissions from a switch to biodiesel enhance the levels of atmospheric oxidants, which increases the oxidation of SO_2 from emissions from all sources. On-road fossil diesel emissions produce nitrate aerosols (Fig. S2c) and contributes 20-40% to the total nitrate aerosol burden over much of Europe. A 10% increase in NO_x emissions are assumed for the replacement of fossil diesel by biodiesel in the B100 case, which results in an increase in the nitrate aerosol burden as shown in Fig. 1c. The production of nitrate aerosols is also affected by the changes in the SO_4, because of the competition for available ammonia. In general, NO_x emissions lead to production of tropospheric O_3 and the on-road fossil diesel sector thus contributes to increased O_3 concentrations (Fig. S2d). The impact on O_3 from a switch to biodiesel is determined by the increase in NO_x, but also by the reductions in CO and VOC emissions. While higher NO_x emissions lead to increased O_3, the reductions in CO and VOC reduces the ozone production. The overall impact in B100 is an increase in the O_3 burden compared to the fossil diesel case (Fig. 1d). Two sensitivity tests with separate perturbations in NO_x and CO+VOC emissions show that increases in NO_x have a stronger impact on O_3 than reductions in CO and VOC.

FIGURE 1

Figure 2 summarizes the global and annual mean RF (relative to a no on-road diesel emissions case) of SLCFs for the current EU on-road fossil diesel sector (FF) and for the sector after the replacement of fossil diesel with biodiesel (B20 and B100). The net RF of SLCFs is positive, mainly determined by the warming of BC. Lund et al.[38] estimated an additional positive RF from BC deposition on snow of 0.3 mW/m^2 (5% of direct BC RF) for the EU on-road fossil diesel sector using the same emissions inventory. The positive RF of O_3 (net of changes due to NO_x/CO/VOC and methane-induced ozone loss) is offset by the negative RF of induced reduction in CH_4. Sulfate, nitrate and organic aerosols give smaller negative contributions. Relative to the FF case, we find reduced global-mean RF of BC, OC and SO_4, a small reduction in RF of O_3 and enhanced forcing from methane and nitrate aerosols in the biodiesel cases. As with surface concentration changes, the change in forcing scales linearly with the emission reductions in the case of the primary aerosols BC and OC and we find a 50% reduction in the positive RF of BC and negative RF of OC for B100 compared to FF. The reduction in the RF of SO_4 is 32% in B100, substantially smaller than the reduction in SO_2 emissions for the reasons discussed above. A 100% replacement of fossil diesel with biodiesel results in a 6% lower O_3 RF and 14% stronger CH_4 forcing. The change in O_3 RF is a combination of the increased O_3 production and enhanced methane-induced loss. The NO_x-induced methane changes can be partly compensated by emissions of CO and VOC. Biodiesel reduces these emissions and thus the compensating effect, which contributes to strengthening the RF of CH_4. Relative to the FF case, a 12% higher RF of nitrate aerosol is found in B100. Similar results are seen in the B20 case for all SLCFs, but with smaller magnitudes due to the smaller emission changes from a 20% blend.

Our simulations show a net positive global annual mean RF of SLCFs from the current EU on-road diesel sector. Given the best estimates of changes in emissions when fossil diesel is replaced by biodiesel the effect of a switch to biodiesel is a reduction in this net warming, from approximately 3 mW/m^2 to 2.8 mW/m^2 in B20 and to 0.7 mW/m^2 in B100. Our calculations do not include forcing due to indirect aerosol effects (IAE) or semi-direct effects, which could affect the results. Some studies have used results from Kvalevåg and Myhre[39] to obtain an estimate of IAE by scaling the direct RF of SO_4.[15,40] With this approach the forcing of IAE due to emissions from the current EU on-road fossil diesel sector is negative and around -1 mW/m^2. However, this approach is highly simplified and does not capture the effect of all aerosol-cloud interactions. For instance, in the case of BC the semi-direct effect is significant due to altered stability of the atmosphere. However, the sign and magnitude of BC semi- plus indirect effect is uncertain.[41,42] Moreover, non-linearities in the response to aerosol perturbations means that the impact on IAE of a switch to biodiesel cannot readily estimated only from changes in emissions.[43,44]

FIGURE 2

Temperature response to SLCFs

Next we show the global-mean temperature response to the SLCFs as a function of time (Fig. 3), again for the current EU on-road fossil diesel sector and for the sector after the replacement of fossil diesel with biodiesel. The left column shows the response to a one year pulse of emissions and the right column shows the response to sustained constant emissions (i.e., a sum of equal pulses). The top panels show the temperature response to aerosols (BC and net of cooling aerosols) and the middle panels show the net of the NO_x/CO/VOC-induced ozone and methane changes. The net of all SLCFs is displayed in the bottom panels. Following the reduced RF, a switch to biodiesel gives a lower net global-mean temperature increase from SLCFs compared to the FF case. This is seen throughout the 80 year period considered and for both pulse and sustained emissions. The changes are mainly driven by the reduced BC warming and stronger net cooling impact of NOx/CO/VOC-induced CH_4 changes and subsequent ozone loss. Because of the longer adjustment time of the latter (approximately 12 years) compared to the other SLCFs, the reduction in net temperature response to SLCFs in B100 relative to the FF case increases over time for sustained emissions. While the absolute values and changes are small, the relative changes are substantial in the sustained B100 case, where the net temperature response is 40% smaller than in the FF case during the first few years, and 80% smaller by year 80. In B20 the temperature change is about 10-15% smaller than in FF. In summary, for the time scales and emissions changes considered here, a reduction in the global-mean climate warming of SLCFs from the EU on-road sector may be obtained from a replacement of fossil diesel with biodiesel.

FIGURE 3

Biodiesel SLCFs versus CO_2

Finally, we examine how the change in SLCFs compares with the impact of changes in the carbon balance resulting from replacing fossil diesel with biodiesel. The global-mean temperature response to EU on-road diesel net SLCFs (from bottom panel of Fig. 3) and CO_2 are compared in Fig. 4, assuming a one year pulse emission (a,c) and sustained constant emissions (b,d). Each individual curve represents the temperature response to either SLCFs or CO_2 from the sector as a whole: Panels a-b and c-d show the temperature responses in the B20 and B100 case, respectively, i.e., after replacing 20% or 100% of the fossil diesel with biodiesel. The different CO_2 biodiesel curves show the temperature response calculated using the different assumptions described in Table 2 and Section S2. In each panel, the temperature response resulting from the EU fossil diesel sector is included for reference (solid blue and black line).

FIGURE 4

The net warming impact of SLCFs is stronger than that from CO_2 in first few years. However, due to the long response time, CO_2 becomes the dominant component over time, as has also been illustrated in previous studies.[15,45] Depending on assumptions for biodiesel blend (i.e., B20 versus B100), additional LUC CO_2 emissions and temporal treatment of the carbon from biomass sources, a broad range in the temperature response to biodiesel CO_2 from the EU on-road sector is calculated. The results reflect the complexity arising from uncertainties in how to account for the net climate impact of CO_2 from biomass. Two features are described in more detail. Firstly, in B20 the C-neutral LCA and IRF_{bio} r5 cases are very similar (Fig. 4a). This is due to the fact that as the assumed rotation period becomes smaller, the fraction of the carbon released by biodiesel combustion is more rapidly sequestered by regrowth and the net biomass carbon emissions hence approaches zero, i.e., “carbon-neutrality”. Secondly, in the IRF_{bio} r50 pulse case there is a temporary longer-term cooling of CO_2 as seen in Fig. 4c. When the IRF_{bio} is used to describe the atmospheric decay of biomass carbon the atmospheric CO_2 fractions becomes temporarily negative due to the uptake of carbon in the various sinks at different timescales, as described in detail in Cherubini et al.[12] The IRF_T of Boucher and Reddy[31] used to calculate temperature response places significant weight on the shorter response timescale of the climate system. Hence, there is insufficient inertia in the system to overcome the cooling induced by this negative CO_2 forcing. The temperature response will depend on the value of the parameters in the IRF_T and these are subject to significant uncertainty.[46] We have performed a sensitivity test using the three alternative IRF_T from Olivié and Peters.[46] Neither of these changes our overall results, however with the IRF_T derived from CMIP5 data the negative temperature responses are smaller and present for a shorter time period.

Figures 4b and d show how the warming of CO_2 accumulates over time in the sustained case under most of the assumptions used here, regardless of whether fossil diesel is replaced by biodiesel. The warming of SLCFs on the other hand reaches a steady-state. Thus, although a switch to biodiesel may under some assumptions result in a lower warming compared to fossil diesel, on-road activity sustained at the present-day level still results in a net climate warming which increases over time. Hence, in addition to biodiesel, significant efficiency improvements, other alternative technologies and/or sustained activity reductions are required to reduce the future climate impact of the EU on-road diesel sector. The exception is the B100 IRF_{bio} case, when a leveling off or even decline in the temperature response to CO_2 is seen (Fig. 4d). This result may overestimate the benefit from biodiesel because the rotation period included in the IRF_{bio} definition only relates to the regrowth of the biofuel feedstock and does not include forest management. Hence, the forest which is assumed to be felled and used for biofuel each year in the sustained case is allowed to continue to grow and capture carbon until 100% regrowth. However, if a rotation period for the management of the forest is considered, the forest could be felled before reaching 100% regrowth, leaving more carbon unsequestered.

The impact of SLCFs likely continues to be small compared to CO_2 from the sector. However, under some assumptions the SLCFs may be relatively more important for the total warming of the sector than in the fossil fuel case. This is found in the specific case where very high biodiesel blend (B100) using feedstock with short rotation periods and low impacts through LUC is assumed. In this case, the warming of SLCFs constitutes over 50% of the total warming of the sector for the first 20 years and 17% after 80 years of sustained emissions. This is significantly higher than in both the other biodiesel cases and the fossil diesel case, where SLCFs provide less than 7% of total warming by year 80. Furthermore, the warming of SLCFs remains higher than that of CO_2 over a longer period compared to the other cases considered. Our results illustrate that improved knowledge of how to account for biofuel CO_2 and LUC impacts is crucial for assessing the net climate impact of biodiesel and relative impacts of SLCFs and CO_2.

Studies suggest significant cooling of climate due to changes in surface albedo resulting from biomass harvesting, especially in regions affected by seasonal snow cover.[13,14,47] This impact is temporary as the albedo gradually reverts during biomass regrowth, with timescale depending on the feedstock rotation period.[35] The temporal behavior of the resulting temperature response if included in our calculations would resemble that due to other SLCFs, i.e., give a substantial initial, but short-lived cooling. However, the strength of the albedo effect depends strongly on harvest region and feedstock.[35]

Discussion

In the case of the current EU on-road diesel sector, our results suggest a reduction in the climate warming from SLCFs if fossil diesel is replaced by biodiesel, based on best estimates of emission changes from existing literature. Although the majority of studies report reduced PM, CO and hydrocarbon emissions and increased NO_x relative to fossil diesel,[15-17,25] there is a significant range in magnitude and some studies also find opposite results.[16] The review by Giakoumis et al.[16] reports changes in NO_x ranging from +60 to -25% for 100% biodiesel blends, and even broader ranges in emission changes for particulate matter (+45 to -80%, majority of estimates show reductions of 20% or more), CO (+90 to -75%, majority of estimates between -20 and -60%) and hydrocarbons (+30 to -100%, majority between -20 and -80%). While changes in the RF of BC and OC scale relatively linearly with emission changes, the net impact on the remaining SLCFs from a switch to biodiesel is more complicated and cannot readily be determined directly from emission changes. Replacing fossil diesel with biodiesel may provide significant co-benefits in terms of air quality due to the reduced PM emissions, but may exacerbate the detrimental effects of NO_x.

This study focuses on the impact of SLCFs following changes in tailpipe emissions and does not account for SLFCs over the entire biodiesel life cycle. It is important to note that there can be significant emissions from various stages in the biodiesel production, which may partly or completely offset the reductions in tailpipe emissions from a switch from fossil diesel.[48-50] For instance, Sheehan et al.[50] report life-cycle reductions of about 30% in PM and CO emissions and 8% in SO_2 from a switch to soybean biodiesel, which is smaller than when only changes at the tailpipe are considered. Furthermore, hydrocarbon emissions increase by more than 30% over the life-cycle despite a strong reduction in tailpipe emissions and the increase in NO_x emissions is enhanced. Significant emissions of NO_2, CO, hydrocarbons and SO_2, particularly at the feedstock cultivation and recovery and fuel production stages, are also found for soybean biodiesel by Delucchi,[48] and comparative LCAs of rapeseed methyl ester reflect increases in NO_x and hydrocarbon emissions compared to fossil diesel.[49,51] Results of LCA of biodiesel differ between studies, feedstocks and even regions.[48-52] Furthermore, the spatial distribution of changes in upstream emissions, and hence the consequent impact on atmospheric concentrations, differs from tailpipe emission changes. Further studies should include a higher level of detail in order to capture a more complete picture of the overall impact.

It is important to note that our results cannot necessarily be directly extrapolated to the use of biofuels in other transport sectors. The global shipping sector gives a net cooling contribution to climate change today, mainly driven by the indirect effect of SO_4 aerosols.[53,54] Righi et al.[20] show that replacing conventional fuel with biofuels in the shipping sector results in a significant decrease in concentrations of SO_4 and hence a reduced cooling climate impact. Depending on the approach used to account for biofuel CO_2 and LUC impacts, the expected switch to a net warming impact of the sector when cooling contributions are reduced and CO_2 accumulates[45] may occur earlier than in the fossil fuel case. In the case of aviation, Krammer et al.[19] show that widespread use of biofuels could result in a scenario where aviation growth is accompanied by flat or decreasing aviation carbon emissions, but an increasing total aviation impact due to contrail-cirrus and other SLCFs. Gasoline vehicles generally have lower emissions of PM than comparable diesel vehicles and a different mix of CO, VOC and NO_x. Hence, the relative effect of replacing gasoline with ethanol can differ significantly from the biodiesel cases of this study.

Furthermore, there can be large temporal and regional differences within the on-road diesel sector. The implementation of strict fuel quality and emission standards has lead to a recent stabilization and decline in EU on-road emissions[55,56] and this reduction is projected to continue in the decades towards 2050, even without biodiesel.[38,57] In this case the advantage in terms of reduced warming of SLCFs from a switch to biodiesel will gradually be reduced over time, which is not accounted for in our sustained emissions case. Outside the OECD countries, less stringent legislation is in place and the fuel sulfur content is higher in many regions.[58,59] Hence, both the magnitude of current emissions and the projected future development differ from that in the EU.[60,61] Furthermore, equal mass emissions in different regions can have different impacts on atmospheric composition and climate, as in the case of ozone precursors.[37,62] An increased use of biodiesel outside the EU could potentially give higher benefits wrt reducing the impact of SLCFs relative to the fossil fuel case, both today and over the near term, and should be studied further. Moreover, it should be noted that a switch to biofuels is of course not the only viable option for reducing the emissions and environmental impact of the transport sector, and alternatives such as an electrification of the vehicle fleet have been proposed to potentially play an equally or more important role.[63-65]

We emphasize that the use of different assumptions for how to account for biomass CO_2 is for illustrative purposes and depends on several simplifications. Firstly, we do make any assumptions about the biodiesel feedstock, but use two factors symmetric around zero for the net CO_2 emissions savings from biodiesel compared to fossil diesel to represent at least a part of the range of possible LUC impacts from existing literature.[66-69] Using any intermediate net saving values in our calculations would produce temperature responses to CO_2 between the responses calculated with the two selected factors. Both higher positive and negative net savings values may be possible, for instance for second-generation biofuels or for large-scale biofuel demands. Secondly, LUC emissions of species other than CO_2 are not considered and it is assumed that the temporal evolution of atmospheric CO_2 from LUC emissions can be represented by the same IRF as for fossil CO_2. Thirdly, LUC impacts are assumed to occur immediately and be constant over time in the sustained emissions case. Further studies should consider more detailed scenarios for replacement of fossil fuels and LUC emissions, as well as activity growth and vehicle fleet development. Finally, in our B100 case the total EU on-road fossil diesel consumption in 2010 is replaced by biodiesel, and we assume that the technical potential and feedstock availability to produce this amount of biodiesel exist.

Given the best estimates for changes in emissions of SLCFs and their precursors when fossil diesel is replaced by biodiesel within the EU, our results show that there is likely to be a reduction in the net positive RF of SLCFs from this sector. However, over time the climate impact of the SLCFs is likely to be small compared to the impacts due to changes in the carbon balance and accompanying LUC under most assumptions. However, in the specific case when biodiesel from feedstock with very short rotation periods and low land-use-change impacts replaces a high fraction of fossil diesel, SLCFs are relatively more important for total warming of the sector than in the other biodiesel cases considered and in the fossil diesel case. These results illustrate the need for improved knowledge of how to account for biofuel CO_2 and LUC impacts in order to assess the net climate impact of biodiesel and relative impacts of SLCFs and CO_2.

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Associated content

Supporting Information Available: Two sections and two figures providing further details about the methodology and additional modeling results. This material is available free of charge via the Internet at http://pubs.acs.org.

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Tables

Table 1: Emissions from the year 2010 EU on-road fossil diesel sector and scaling factors applied in the biofuel cases.

Case	**BC**	**CO**	**NH3**	**NOx**	**OC**	**SO2**	**VOC**
Fossil diesel [kt]	**113**	**1045**	**9**	**3863**	**25**	**13**	**177**
% change B100	-50	-50	-	+10	-50	-90	-65
% change B20	-10	-10	-	+2	-10	-20	-20

Table 2: Summary of assumptions about how to account for CO_2 from biodiesel used in the temperature response calculations. A detailed description can be found in Section S2.

Case	Description
Fossil CO_2	Fossil diesel case, included for reference
C-neutral LCA	Biodiesel is assumed carbon neutral (conventional LCA), i.e., no net CO2 emissions
Low LUC	Assuming a net CO_2 emission saving from biodiesel relative to fossil diesel of 25 g CO_2/MJ[1]
High LUC	Assuming a net CO_2 emission saving from biodiesel relative to fossil diesel of -25 g CO_2/MJ[1]
IRF_{bio} r5	Atmospheric CO_2 concentration from biodiesel calculated using specific biogenic IRF[2], assuming feedstock rotation period 5 years
IRF_{bio} r50	Atmospheric CO_2 concentration from biodiesel calculated using specific biogenic IRF[2], assuming feedstock rotation period 50 years

[1] Net savings equals direct saving from consumption of biodiesel relative to fossil fuel minus additional emissions from land-use change impacts. These two cases are very loosely based on results from Laborde[68], but the symmetric value of ±25 gCO_2/MJ is selected to reflect a larger range of possible LUC impacts, rather than a specific biofuel feedstock.

[2] Cherubini et al.[12]

Figure captions

Figure 1: Modeled change in atmospheric burden of (a) BC, (b) SO_4, (c) nitrate aerosol and (d) ozone resulting from changes in emissions when fossil diesel is replaced by biodiesel in the B100 case. Units [$\mu g/m^2$] ([10^{-2} DU] for ozone).

Figure 2: Global and annual mean radiative forcing due to SLCFs for sustained year 2010 EU on-road fossil diesel emissions (FF) (relative to a no on-road diesel emissions case), and for sustained year 2010 EU on-road diesel emissions after a 20% (B20) and 100% (B100) replacement of fossil diesel by biodiesel.

Figure 3: Global-mean temperature change due to SLCFs from the current EU on-road fossil diesel sector (FF) and from the sector after a 20% (B20) or 100% (B100) replacement of fossil diesel with biodiesel. Left panels show the response to pulse emissions and right panels show the impact of sustained current emissions. Pink lines = BC, red lines = net of sulfate, nitrate and organic carbon aerosols, green lines = net of NOx/CO/VOC-induced changes in ozone and methane and blue lines = net of SLCFs.

Figure 4: Global-mean temperature responses to net SLCFs and CO_2 from the EU on-road diesel sector, assuming a one year pulse emission (left) and sustained constant emissions (right). Each individual curve represents the temperature response to either SLCFs or CO_2 from the current EU on-road sector as a whole, in our fossil diesel and biodiesel cases. In each panel, the temperature response resulting from the EU fossil diesel emissions is included for reference (solid blue and black line). The top and bottom panels show the temperature responses to emissions from the sector in the B20 and B100 case, respectively, i.e., after a 20% or 100% replacement of fossil diesel by biodiesel. The different CO_2 biodiesel curves show the temperature response calculated using the different assumptions for how to account for the biodiesel CO_2 and land-use change (LUC) impacts (described in Table 2 and Section S2).