



RESEARCH ARTICLE

Lifetimes, direct and indirect radiative forcing, and global warming potentials of ethane (C₂H₆), propane (C₃H₈), and butane (C₄H₁₀)

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The atmospheric abundance of the non-methane volatile organic compounds (NMVOCs) ethane, propane, and butane increased during the industrial era. In addition to weak absorption and emission of longwave radiation, these gases influence the atmospheric radiative balance indirectly, mainly as precursors for ozone (O₃), and through reaction with the hydroxyl radical (OH), which leads to less OH and thereby longer atmospheric lifetime of methane (CH₄). In this study, we have calculated lifetimes, direct and indirect radiative forcing (RF), and global warming potentials (GWPs) for the three compounds, using a self-consistent methodology. Results show net RF per unit emission of 1.0, 0.9, and 0.6 mW m⁻² (Tg year⁻¹)⁻¹ for ethane, propane, and butane, respectively. For all compounds, the direct effect is considerably smaller than the indirect effects (6% or less of the total). The indirect O₃ and CH₄ effects are approximately of the same magnitude. Net GWPs for a 100-year time horizon are 10 for ethane and propane, and 7 for butane, whereof the direct GWPs are <1 for all compounds. The net GWPs are generally higher than previous estimates, mainly because our calculations include emissions for a full year rather than one season. For the compounds studied here, 100-year GWP values do not differ substantially between each compound, considering the large uncertainties involved, and this may indicate that using values representative for a lump of NMVOCs may be sufficient. However, the climate effects may differ more between NMVOC compounds other than alkanes, such as alkenes and aromatics.

KEYWORDS

ethane, propane, butane, radiative forcing, GWP

1 | INTRODUCTION

Since preindustrial times, atmospheric ethane and propane concentrations in the Northern Hemisphere have increased by a factor of 3–4, mainly due to fossil fuel emissions (Nicewonger et al., 2016). After a period of decline between the 1970s and year 2005 (Simpson et al., 2012; Helmig et al., 2016), ethane concentrations have over the last decade

picked up again (Franco et al., 2016; Helmig et al., 2016). Ethane, propane, and butane are classified as non-methane hydrocarbons (NMHCs), which are part of a larger group of compounds named non-methane volatile organic compounds (NMVOCs), and ethane is the NMHC with the longest lifetime and highest abundance. Together with propane, ethane is also a compound where atmospheric chemistry models generally underestimate present-day concentrations

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(Thompson et al., 2003; Stein and Rudolph, 2007; Emmons et al., 2015), likely due to underestimation of emissions (Franco et al., 2016; Dalsøren et al., 2018).

NMVOCs influence radiative forcing (RF) of climate change through several mechanisms. First, several NMVOCs have a direct influence on climate as they act as modest greenhouse gases (Highwood et al., 1999; Sihra et al., 2001). Second, and more importantly, NMVOCs influence climate indirectly through reaction with the hydroxyl radical (OH), and this leads to increased production of ozone (O₃) and longer lifetime of methane (CH₄) (Collins et al., 2002; Fry et al., 2012; Fry et al., 2014). Ozone production is also increased in a longer-term because methane is a precursor for ozone, and this is known as the primary mode effect (Wild and Prather, 2000; Wild et al., 2001; Stevenson et al., 2013).

Indirect climate effects can vary significantly between different NMVOCs, for example, due to different potentials to produce ozone (Derwent et al., 1998, 2001). However, only one study (Collins et al., 2002) has calculated indirect climate effects of several NMVOCs separately. Here, we calculate lifetimes, and both direct and indirect climate effects of ethane, propane, and butane, to reflect recent developments in models and methodology. Global warming potentials (GWPs), which are of interest to the scientific community, policy-makers, and industry, are provided for each of the three compounds.

2 | METHODS

To estimate the direct radiative efficiencies, defined as the RF per increase in atmospheric concentration, we use the Hodnebrog et al. (2013) method, which is an updated version of the Pinnock et al. (1995) method. With these methods the instantaneous radiative efficiency is calculated directly when the experimental absorption cross section of the gas is known. Experimental absorption spectra in this study are from the freely available online PNNL database (Sharpe et al., 2004), measured at sample temperatures of 298 K. In line with Hodnebrog et al. (2013), we account for stratospheric temperature adjustment by adding 10% of the instantaneous radiative efficiency. The method for calculating direct radiative efficiencies assumes the gas to be well mixed in the atmosphere and, especially for short-lived compounds, this radiative efficiency needs to be modified to account for a more realistic atmospheric distribution. Hodnebrog et al. (2013) explored this issue by performing an extensive set of chemistry-transport model (CTM) and radiative transfer model (RTM) simulations for compounds with a wide range of atmospheric lifetimes, with the only loss being reaction with OH in the troposphere, and with an assumed emission distribution as an industrial compound. Based on this they derived so-called lifetime correction factors, which are, for a given lifetime, the ratio in annual mean RF between an RTM simulation using monthly mean atmospheric

distributions from a CTM, and an RTM simulation using a constant global and annual mean vertical profile (with the global and annual mean surface mixing ratio being the same in the two simulations). Since the hydrocarbons in this study also are lost through reaction with OH and mainly are from anthropogenic sources, we believe these lifetime correction factors give a reasonable approximation to account for a heterogeneous atmospheric distribution in the atmosphere. Applying the lifetime correction factors leads to a reduction in the radiative efficiency of a compound (see figure 9 and equation 2 in Hodnebrog et al. (2013)), mainly because a shorter lifetime leads to a lower atmospheric burden.

The OsloCTM3 model (Søvde et al., 2012) is used to calculate changes in concentrations following a doubling of anthropogenic and natural emissions for each of ethane, propane, and butane, in three different simulations. The reference simulation is first spun up for 1 year, and then the reference and perturbation simulations have been run for 500 days, whereof the latter 365 days are analyzed. The reference simulation uses emissions and meteorological input data for the year 2008. Year 2008 anthropogenic emissions of CO, NO_x, NH₃, sulfur, and NMVOCs are from the EDGAR v4.2 inventory (EC-JRC/PBL, 2011). The EDGAR v4.2 inventory only provides total NMVOC emissions per sector (e.g., transportation, industry) and disaggregation into individual species (e.g., ethane, propane) emissions was based on the emissions reported by (Lamarque et al., 2010). For biomass burning emissions we use GFEDv3 (van der Werf et al., 2010) year 2008 emissions. The parametrization of lightning NO_x emissions are described in Søvde et al. (2012). For NO_x from soil, and CO and NMVOCs from the oceans, we use the year 2000 emissions in the RETRO inventory (Schultz et al., 2008). Sources for natural sulfur emissions are described in Berglen et al. (2004). The emissions from vegetation of CO and NMVOCs are from the MEGAN-MACC year 2008 inventory (Sindelarova et al., 2014). For natural NH₃ sources we use emissions from Bouwman et al. (1997) for 1990. The model uses 3-hourly meteorological forecast data from the European Centre for Medium-Range Weather Forecasts (ECMWF) Integrated Forecast System (IFS) (see Søvde et al. (2012) for details). A coupled tropospheric and stratospheric version of OsloCTM3 is run with 109 species involved in atmospheric ozone, methane, and NMVOC chemistry. The model includes the emissions, photochemistry, and degradation products of the major NMHCs (C₂H₆, C₃H₈, C₄H₁₀, C₆H₁₄, C₂H₄, C₃H₆, C₄H₁₀, m-xylene, and isoprene) and major NMVOCs (e.g., acetone, formaldehyde, and acetaldehyde). Formation of long-lived reservoir compounds like PAN is thereby accounted for. Other NMHCs are not treated explicitly, but if significant, their emissions are assigned to one of the species listed above (see table A1 in Søvde et al. (2008) for a complete list of NMHCs). They are assigned to a NMHC that produces the same number of RO₂ radicals (thus determining the potential for ozone production) and has a similar lifetime (thus

affecting the same region). To calculate O₃ RF including stratospheric temperature adjustment, monthly mean ozone concentration fields from the CTM simulations are fed into the Oslo RTM (Myhre et al., 2011), which consists of a broadband code for longwave radiation (Myhre and Stordal, 1997), and a multistream DISORT code for shortwave radiation (Stamnes et al., 1988).

Methane concentrations in the OsloCTM3 simulations are prescribed throughout the atmosphere based on monthly mean methane concentrations for the year 2008 from the Dalsoren et al. (2016) methane study. The CH₄ lifetime in the reference simulation is 9.7 years, which is the same as the mean of 16 chemistry-climate models and close to the observation-derived estimate of 10.2 years for the year 2000 in Naik et al. (2013). To calculate CH₄ RF, the change in OH between the reference and perturbation simulations is used to calculate the change in CH₄ lifetime ($\Delta\tau_{\text{CH}_4} = \tau_{\text{CH}_4,\text{pert}} - \tau_{\text{CH}_4,\text{ref}}$). The relative change in CH₄ lifetime is then used to calculate, for each perturbation, the ppb difference in CH₄ from a background concentration ($\Delta\text{CH}_4 = \Delta\text{CH}_4_{\text{background}} \times f \times [\Delta\tau_{\text{CH}_4}/\tau_{\text{CH}_4,\text{ref}}]$). To account for the impact of CH₄ changes on its own lifetime, a feedback factor $f = 1.34$ was applied. The CH₄ RF is calculated with the same methodology as in IPCC AR5 (Myhre et al., 2013), but with some recent updates. In IPCC AR5 a background CH₄ concentration ($\Delta\text{CH}_4_{\text{background}}$) of 1803 ppb and a radiative efficiency of 0.363 mW m⁻² ppb⁻¹ were used. The CH₄-induced O₃ RF (primary mode; hereafter denoted CH_{4(O3)} RF) was assumed to be a factor of 0.5 times the CH₄ RF (Myhre et al., 2013), and the impact of CH₄ changes on stratospheric water vapor (hereafter denoted CH_{4(SWV)} RF) was assumed a factor 0.15 times the CH₄ RF (Myhre et al., 2007). The CH_{4(O3)} RF and CH_{4(SWV)} RF are calculated as described above, but for the radiative efficiency of CH₄ the recent update from Etminan et al. (2016), which included shortwave absorption of CH₄, is adopted. For a background CH₄ concentration of 1834 ppb as for 2015, the radiative efficiency is 0.443 mW m⁻² ppb⁻¹, which is 22% higher than the IPCC AR5 value.

The GWP is “the time-integrated commitment to climate forcing from the instantaneous release of 1 kg of a trace gas expressed relative to that from 1 kg of carbon dioxide” (IPCC, 1990), and is calculated here for a 100-year time horizon. Details about equations and parameters for calculating direct and indirect GWPs are given in Hodnebrog et al. (2013) and Aamaas et al. (2013), respectively, and are

consistent with the methodology in IPCC AR5 (Myhre et al., 2013). Collins et al. (2002) have included the effect of NMVOCs oxidizing to CO₂. This effect is not included here because it is often accounted for in CO₂ inventories, but, if included, would have increased the GWP estimates by about 3.67 (Fry et al., 2014).

3 | RESULTS

Table 1 shows that the direct radiative efficiencies calculated in this study compare relatively well with previous literature (<20% difference) when we assume a constant vertical and horizontal distribution in the atmosphere. The slightly lower values in Sihra et al. (2001) can partly be explained by their integrated absorption cross sections, which were lower than in the PNNL data used here for all three compounds (Table S1, Supporting Information). Due to the short lifetimes (ranging from ~1 week to ~2 months), one should account for the fact that the compounds are not well mixed in the atmosphere and therefore the RF is reduced (see section 2). Table 1 shows that applying the lifetime correction factors from Hodnebrog et al. (2013) leads to substantially lower radiative efficiencies.

In Table 2, the direct radiative efficiency is converted to specific RF, for comparison to indirect effects, which are commonly given as the RF per unit emission. While radiative efficiency is given here as the RF per unit increase in concentration with units of mW m⁻² ppb⁻¹, the specific RF is given as the RF per unit increase in emissions with units of mW m⁻² (Tg year⁻¹)⁻¹. The direct radiative efficiencies are converted to specific RF by using the perturbed emissions and surface concentrations from the OsloCTM3 simulations for each of the three compounds. From Table 2 it is evident that the lifetime-corrected direct effect is small compared to the indirect effects, particularly for propane and butane. The main reason is seen in Figure 1, which shows the absorption cross sections on top of the radiative efficiency curve derived in Hodnebrog et al. (2013). While the atmospheric absorption is most efficient in the so-called atmospheric window region, approximately in the range 800–1200 cm⁻¹, all three compounds absorb only weakly in this region and have their main infrared absorption band around 1500 cm⁻¹.

For all compounds, the short-term ozone effect and the long-term methane effect are of comparable magnitude in

TABLE 1 Lifetimes and direct radiative efficiencies

	τ (days)	Direct radiative efficiency (mW m ⁻² ppb ⁻¹)			
		Sihra et al. (2001), constant profile	Highwood et al. (1999), constant profile	This study, constant profile	This study, lifetime corrected
Ethane	58	3	3.2	3.5	1.2
Propane	13	3	3.1	3.0	0.36
Butane	6.8	4	4.7	4.1	0.28

TABLE 2 Specific radiative forcing ($\text{mW m}^{-2} [\text{Tg year}^{-1}]^{-1}$)

	Direct	O ₃	CH ₄ (net) ^a	Aerosols	Total	Reference
This study (per VOC)						
Ethane	0.056	0.36	0.54		0.95	
Propane	0.0061	0.37	0.50		0.88	
Butane	0.0030	0.30	0.30		0.60	
Previous studies (per VOC)						
NMVOIC		0.23	0.25		0.49	Stevenson et al., (2013)
This study (per C)						
Ethane	0.070	0.45	0.68		1.2	
Propane	0.0074	0.45	0.62		1.1	
Butane	0.0037	0.36	0.36		0.72	
Previous studies (per C)						
NMVOIC		0.31	0.21	-0.04	0.48	Fry et al. (2012) ^b
NMVOIC					0.21	Fry et al., (2014) ^c
NMVOIC		0.77	0.64	-0.40	1.00	Bellouin et al. (2016) ^d

^a Includes effects on primary mode O₃ (CH₄(O₃) RF) and (for this study and Bellouin et al. (2016)) stratospheric water vapor (CH₄(SWV) RF). In Fry et al. (2012), CH₄(O₃) RF is included in the O₃ term and not the CH₄ term.

^b Anthropogenic emissions were perturbed for East Asia, Europe, North America, and South Asia.

^c Net RF from changes in O₃, CH₄, and SO₄ (direct effect) accounted for in the total, but the split between the different terms is not given in the paper.

^d Numbers here are averages of results for summer and winter perturbations.

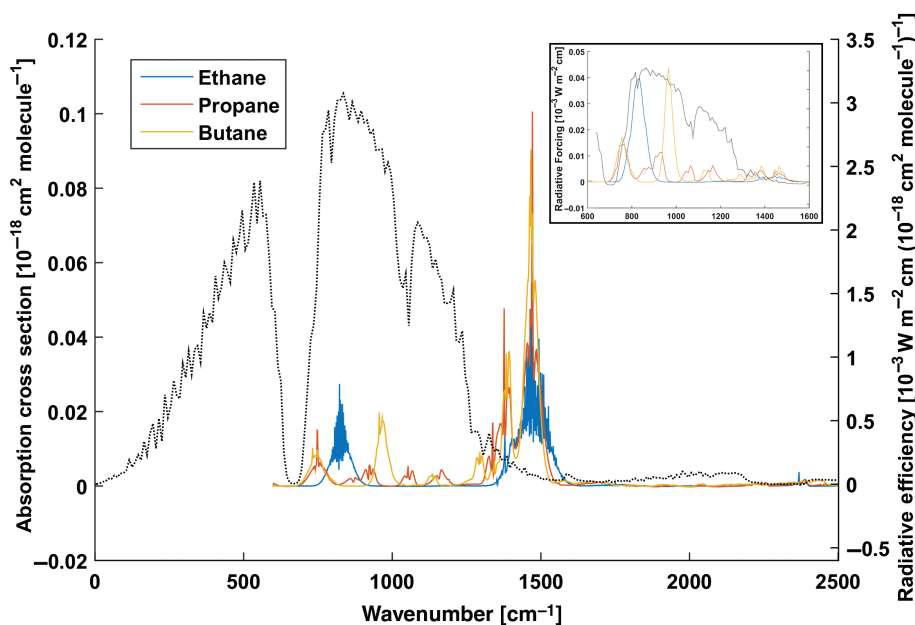


FIGURE 1 Absorption cross-sections of ethane, propane, and butane from PNNL (Sharpe et al., 2004; solid lines, left y-axis), and the radiative efficiency curve derived in Hodnebrog et al. (2013) (dotted line, right y-axis). The colored lines in the inset figure show the two curves multiplied, that is, the radiative forcing per wavenumber for each compound, zoomed in for the wavenumber range 600–1600 cm^{-1}

terms of specific RF. Using the updated numbers for CH₄ from Etminan et al. (2016) increases the long-term methane term by 22% compared to using the IPCC AR5 methodology (Table 2; Table S2). Results from a simulation where ethane emissions only in the Arctic region (north of 60°N) are perturbed, show almost unchanged long-term methane effect, but a 30% lower short-term ozone effect compared to perturbing global ethane emissions (Table S3; Table 2). For ozone, this result is consistent with the less active photochemistry at high latitudes. The almost unchanged methane effect implies that the integrated CH₄ + OH flux is similar in the two experiments, and may be explained by the strong temperature dependence of the CH₄ + OH reaction rate. In

the Arctic ethane experiment, high biomass burning emissions lead to a large fraction of the emissions occurring during summer, when temperatures are higher. Ethane in the global experiment is, particularly in the tropics, much more effectively transported to high altitudes, where temperatures are much lower than at the surface. In total, these effects contribute toward similar mean temperatures for the CH₄ + OH reaction in the two experiments, and could partly explain the similar long-term methane effect. However, differences in where OH is located, which is mainly controlled by the H₂O distribution, are also factors contributing to the integrated CH₄ + OH flux. For an Arctic ethane emission source with a less pronounced seasonal variation, the long-

term methane effect is expected to be weaker than for a global emission perturbation.

Specific RFs of ethane, propane, and butane from this study are also given on a per Tg[C] basis, for comparison to NMVOCs from previous studies (Table 2). Generally, our values are higher than previous estimates, but it should be kept in mind that different NMVOC species have different properties (e.g., potentials to produce ozone) and hence, results for individual NMVOCs (e.g., ethane) are not directly comparable to a lump of NMVOCs. In addition, while we have chosen to perturb all emissions (anthropogenic, natural, and biomass burning), other studies have perturbed anthropogenic emissions only. Table 2 shows that our results compare relatively well with the multimodel studies of Stevenson et al. (2013) and Bellouin et al. (2016). Results from Bellouin et al. (2016) also show that the influence of NMVOCs on aerosols (direct and indirect aerosol effects) could offset a large part of the ozone and methane contributions, but the three models involved strongly disagree on the sign and magnitude of the aerosol term. The results of Fry et al. (2012, 2014) are lower. One reason may be that the results of Fry et al. (2012) are based on four emission regions, and not a global emission perturbation. Fry et al. (2014) attribute their low result to greater NMVOC/NO_x emission ratios compared to previous studies, and this would lead to less sensitivity to NMVOC emissions.

TABLE 3 Global warming potentials for a 100 year time horizon

	Direct	O ₃	CH _{4(net)} ^a	Aerosols	Total	Reference
This study (per VOC)						
Ethane	0.39	3.9	5.9		10.2	
Propane	0.018	4.0	5.5		9.5	
Butane	0.0055	3.3	3.2		6.5	
Previous studies (per VOC)						
Ethane		2.6	2.9		5.5	Collins et al. (2002) ^b
Propane		0.6	2.7		3.3	Collins et al. (2002) ^b
Butane		1.7	2.3		4.0	Collins et al. (2002) ^b
Ethane	<1					Wallington et al. (2015)
Propane	<1					Wallington et al. (2015)
Butane	<1					Wallington et al. (2015)
This study (per C)						
Ethane	0.49	4.9	7.4		12.8	
Propane	0.022	5.0	6.7		11.7	
Butane	0.0067	3.9	3.9		7.8	
Previous studies (per C)						
NMVOC		3.6	2.4	-0.4	5.5 (±2.1)	Fry et al. (2012) ^c
NMVOC					2.36	Fry et al. (2014) ^d
NMVOC		8.4	6.9	-4.4	10.9 [-2.5, 29]	Aamaas et al. (2016) ^e

^a Includes CH_{4(O3)} RF and (for this study and Aamaas et al. (2016)) CH_{4(SWV)} RF. In Fry et al. (2012), CH_{4(O3)} RF is included in the O₃ term and not the CH₄ term.

^b They include also effect on CO₂, but this is not included in the total here for comparability.

^c Anthropogenic emissions were perturbed for East Asia, Europe, North America, and South Asia. The uncertainty is the SD based on 11 models.

^d GWP due to changes in O₃, CH₄, and SO₄ (direct effect) accounted for in the total, but the split between the different terms is not given in the paper.

^e Numbers here are averages of results for summer and winter perturbations. Results in Aamaas et al. (2016) are from model simulations in Bellouin et al. (2016). The range given is the minimum and maximum value based on four models.

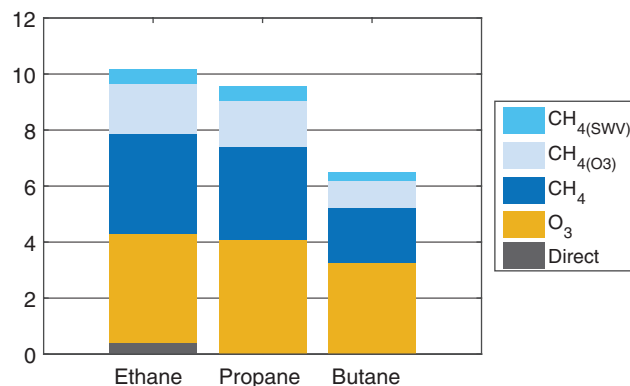


FIGURE 2 GWP for a 100-year time horizon (given on a per kg [VOC] basis) for each of the three compounds, separated into contributions from direct and indirect effects

GWP values for a 100-year time horizon are presented in Table 3 and Figure 2. Our values agree with Wallington et al. (2015) who estimated 100-year GWPs for the direct effect for all three compounds to be <1 when accounting for the fact that short-lived compounds are not well-mixed in the atmosphere. To our knowledge, only one 3-D CTM study (Collins et al., 2002) has presented indirect GWPs for separate NMVOC species. For all of ethane, propane, and butane, our GWP 100 year values are higher than in Collins et al. (2002) (Table 3). Although different models are likely to give different results (Stevenson et al., 2013), most

likely, these differences are to a large degree caused by different experimental designs. While we perturbed annual emissions of each species, the perturbation in Collins et al. (2002) was done for January. Since a major part of NMVOC emissions occur in the Northern Hemisphere, the ozone produced is likely to be lower during January when the photochemistry is less active. In addition, Collins et al. (2002) found that the different PAN chemistry of the NMVOC breakdown products was very important in determining GWPs of individual NMVOCs, and that this resulted in lower O₃ RF for propane compared to ethane and butane. Different NMVOC oxidation chemistry is therefore also a likely factor explaining differences in GWP values in our study compared to Collins et al. (2002).

4 | CONCLUSIONS

We have used an up-to-date and self-consistent methodology to calculate lifetimes, RF and GWPs for three of the most abundant NMVOCs in the atmosphere. The direct radiative efficiency for ethane, propane, and butane are lower than earlier studies because we account for heterogeneous distribution in the atmosphere for these compounds. Specific RF for the indirect effects, through influences on ozone and methane, are considerably larger than the direct effect, and generally higher than estimated in previous studies. Calculated GWPs for a 100-year time horizon are 10, 10, and 7 for ethane, propane, and butane, respectively, and higher than in an earlier study (Collins et al., 2002) where emissions were perturbed during January only.

It is important to stress that GWPs for short-lived species are uncertain since they have a heterogeneous distribution in the atmosphere, and GWPs therefore vary widely depending on assumptions of season (Aamaas et al., 2016; Bellouin et al., 2016) and region (Fry et al., 2012; Fry et al., 2014; Aamaas et al., 2016; Bellouin et al., 2016) for the emitted compound. Hodnebrog et al. (2013) estimate 23% uncertainty in the direct RF for short-lived compounds. In Bellouin et al. (2016), the inter-model relative *SD* for the indirect ozone and methane RF for NMVOC is 44%. For GWP values, uncertainties in the absolute GWP for CO₂, and in the compounds' lifetime, must also be considered. Considering the large uncertainties involved, 100-year GWP values for the alkanes studied here may not differ significantly from the more common approach of using values from a lump of NMVOCs. However, this conclusion may not hold for other NMVOCs, such as alkenes and aromatics.

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SUPPORTING INFORMATION

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