Supplementary Information

Global modeling of trifluoroacetic acid surface concentration and deposition from the gas-phase oxidation of a wide range of precursor hydrofluoroolefins

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Kinetic studies of the reaction of OH radicals and O_3 with HFOs studied

When undertaking kinetic studies, the technique used may have a bearing on the results obtained, where direct determinations (e.g. flash-photolysis with Laser-induced fluorescence detection of the OH radical) are favoured over indirect studies such as relative rate studies which itself relies on accurate kinetic parameters for the reference compound or compounds that are used to compare loss rates with the compound of interest. It also emerges that purity of sample will play a role, reactive impurities in the sample can lead to erroneous lost

(increased) rates being returned and hence the rate constant retrieved. In general, experimental studies are preferred to theoretical ones and some example comparisons show why this is so. There are several proposed HFOs in addition to HFO-1234yf, that could produce TFA on oxidation. In this supplementary section, we review the kinetic determinations for their reaction with OH radicals and O_3 and where needed justify the rate coefficient data used in the main study. In 2015, Volmer et al.[1] reported the first observations in the atmosphere of HFO-1234yf ($CF_3CF=CH_2$ starting in 2012) and HFO-1234zeE ($CF_3CH=CHF$ starting in 2011) at a clean air and polluted site in Switzerland. Atmospheric observations of these compounds were also reported in Germany[2] in subsequent years and in 2023[3] observations of HFO-1336mzzmZ ($CF_3CH=CHCF_3$) were also made in Switzerland. Therefore, other HFOs may already be used but these three HFOs have been observed.

For the 15 HFOs considered in this study that all contain at least one CF_3 group we review gas-phase kinetic studies in section 1. We collect estimates of their lifetime, GWP, and potential yield of TFA and these data are summarised in table S1. In section 1 a discussion of kinetic determinations and why a kinetic parameterisation has been chosen for this study is presented.

Section 1: Kinetic data for the reaction between the HFO and OH radicals and O_3.

HFO-1234yf $CF_3CF=CH_2$

Rate constant (OH) / cm^3 $molecule^{-1}$ s^{-1}	**Temp range/K**	**Ref**
1.05×10^{-12}	296	Nielsen et al.[4]
14.1×10^{-13} exp(-64/T)	252-370	Orkin et al.[5]
1.145×10^{-12} exp(-13／T)	220-298	Orkin et al.[6]
4.06×10^{-13} (T／298)$^{1.17}$exp(296／T)	298-370	Orkin et al.[6]
1.26×10^{-12}exp(-35/T)	**206-380**	**Papadimitriou et al.[7]**
1.54×10^{-12} exp (-100/T)	250-430	Tokuhashi et al.[8]

Rate constant (O_3) / cm^3 $molecule^{-1}$ s^{-1}	**Temp range/K**	**Ref**
2.77×10^{-21}	**296**	**Nielsen et al.[4]**
2.56×10^{-21}	296	McGillen et al.[9]

It is interesting to note that the difference between the two determinations by Orkin et al.,[5,6] is due to impure samples in the 1997 study[5] compared with that from 2010[6] and that those impurities affected the kinetic analysis. All temperature dependent studies[6,7,8] show Arrhenius behaviour with the most recent determination by Tokuhashi et al.[8] having the strongest temperature dependence (smallest rate constant at low temperatures). The estimated lifetime is around 12 days and a GWP of around 4 (see table S1). The work of Papadimitriou et al.[7] has been chosen because the temperature range (206-380 K) investigated is the widest and the percentage difference between all the studies [6,7,8] is small over the temperature ranges studied, i.e. 6% difference between Papadimitriou et al.[7] and Tokuhashi et al.[8] at 250 K and less than 2% over the whole temperature range for the studies of Papadimitriou et al.[7] and Orkin et al.[6] There are two reported studies for the rate constant for the reaction between the HFO and O_3[4,9] for which there is no significant difference within the combined uncertainties. Since we adopt the work of Nielsen and co-workers for other HFO rate coefficients with ozone we have adopted the study of Nielsen et al.[4]

HFO-1225yeZ (CF$_3$CF=CHF)

Rate constant (OH) / cm^3 molecule^{-1} s^{-1}	**Temp range/K**	**Ref**
1.22×10^{-12}	296	Hurley et al.[10]
7.30×10^{-13}exp(165/T)	**206-380**	**Papadimitriou et al.[7]**

Rate constant (O_3) / cm^3 molecule^{-1} s^{-1}	**Temp range/K**	**Ref**
1.45×10^{-21}	**296 K**	**Hurley et al.[10]**

A temperature dependent study by Papadimitriou et al.[7] shows Anti-Arrhenius behaviour with an estimated lifetime of 10 days. A room temperature study[9] is reported that is in agreement with Papadimitriou et al.[7] The lifetime of 10 days estimated by Papadimitriou et al.[7] is in contrast with the 18 days estimated by Hurley et al.[10] based on room temperature data only. Two studies that looked at the mixture of stereoisomers[11,12] are noted but not considered further. The temperature dependent kinetic data from Papadimitriou et al.[7] are used in this study for the reaction with OH radicals. The only study[10] provides a rate coefficient for the reaction with O_3.

HFO-1225yeE (CF$_3$CF=CHF)

Rate constant (OH) / cm^3 molecule^{-1} s^{-1}	**Temp range/K**	**Ref**
2.15×10^{-12}	**296**	**Hurley et al.[10]**

Rate constant (O_3) / cm^3 molecule^{-1} s^{-1}	**Temp range/K**	**Ref**
1.98×10^{-20}	**296**	**Hurley et al.[10]**

Mixed isomers rate constants with OH

HFO-1225ye(E/Z)	2.62×10^{-12} (298 K)	Tovar et al.[11]
HFO-1225ye(E/Z)	7.85×10^{-15} exp(1765/T)	Rivela et al.[12]

Only a room temperature study[10] is reported for the reaction with OH radicals and a lifetime of 10 days is estimated. The same study[10] provides a rate coefficient for the reaction with O_3.

HCFO-1233zdE (CF$_3$CH=CHCl)

Rate constant (OH) / cm^3 molecule^{-1} s^{-1}	**Temp range/K**	**Ref**
1.14×10^{-12} exp(-330/T)	**213-376**	**Gierczak et al.[13]**
7.20×10^{-13} exp(-237/T)	220-370	Orkin et al.[14]
4.4×10^{-13}	295	Andersen et al.[15]
3.61×10^{-13}	295	Andersen et al.[16]

Rate constant (O_3) / cm^3 $molecule^{-1}$ s^{-1}	**Temp range/K**	**Ref**
1.46×10^{-21}	**295**	**Andersen et al.[15]**

There are two temperature dependent studies that are in good agreement over the temperature range.[13,14] Following previous determinations we have used the work of Gierczak et al.[13] as it was determined over a slightly larger temperature range and that it is the only study for the corresponding Z isomer. In addition, there is a room temperature determination[15] that is 20-40% larger than the two temperature dependent studies, on correction for Cl atom chemistry the room temperature rate constant was reduced.[16] There is one study for the reaction with ozone.[15]

HCFO-1233zdZ ($CF_3CH=CHCl$)

Rate constant (OH) / cm^3 $molecule^{-1}$ s^{-1}	**Temp range/K**	**Ref**
$7.22 \times 10^{-19} T^2 \exp(800/T)$	**218-376**	**Gierczak et al.[13]**
8.45×10^{-13}	295	Andersen et al.[16]

Rate constant (O_3) / cm^3 $molecule^{-1}$ s^{-1}	**Temp range/K**	**Ref**
1.53×10^{-21}	**295**	**Andersen et al.[16]**

There is one temperature dependent study[13] and Gierczak and co-workers note anti-Arrhenius behaviour compared with the corresponding E isomer, a room temperature study is in reasonable agreement with the temperature dependent study.[16] There is one study for the reaction with ozone.[16]

HFO-1234zeE ($CF_3CH=CHF$)

Rate constant (OH) / cm^3 $molecule^{-1}$ s^{-1}	**Temp range/K**	**Ref**
$7.6 \times 10^{-13}(T/298)^{2.44}\exp(666/T)$	263-358	Antinolo et al.[17]
$1.115 \times 10^{-13}(T/298)^{2.03}\exp(+552/T)$	**220-370**	**Orkin et al.[6]**
$1.05 \times 10^{-12}\exp(-118/T)$	253-328	Zhang et al.[18]
$4.86 \times 10^{-13}(T/298)^{0.727}\exp(110/T)$	250-430	Tokuhashi et al.[19]
9.25×10^{-13}	295	Sondergaard et al.[20]

Rate constant (O_3) / cm^3 $molecule^{-1}$ s^{-1}	**Temp range/K**	**Ref**
2.81×10^{-21}	**295**	**Sondergaard et al.[20]**

There are several temperature-dependent studies[6,17,18,19] with all in close agreement. We have adopted the study by Orkin et al.[6] given its wide temperature dependence and lowest temperature studies. The room temperature work of Sondergaard et al.[20] is higher than the temperature dependent studies. There is only one study for the reaction with ozone.

HFO-1234zeZ ($CF_3CH=CHF$)

Rate constant (OH) / cm^3 $molecule^{-1}$ s^{-1}	**Temp range/K**	**Ref**
$1.0 \times 10^{-12}\exp(61/T)$	263-358	Antinolo et al.[17]
$9.11 \times 10^{-13}\exp(114/T)$	**253-328**	**Zhang et al.[18]**

$4.55 \times 10^{-13}(T/298)^{0.367}\exp(350/T)$	250-430	Tokuhashi et al.[19]
1.2×10^{-12}	295	Nilsson et al.[21]

Rate constant (O_3) / cm^3 molecule^{-1} s^{-1}	**Temp range/K**	**Ref**
1.65×10^{-21}	**295**	**Nilsson et al.[21]**

There are three temperature dependent studies that all show Anti-Arrhenius behaviour but the rate constants derived from the work of Tokuhashi et al.[19] is significantly larger than the other two. We have adopted the work of Zhang et al.[18] as it covers a slightly lower temperature range, noting that the difference between the two studies[17,18] is between 8-14% over the tropospheric temperature range. The room temperature study of Nilsson et al.[21] is in good agreement with Antinolo et al.[17] and Zhang et al.[18]. There is one study of the reaction with ozone.[21]

HFO-1336mzzE $CF_3CH=CHCF_3$

Rate constant (OH) / cm^3 molecule^{-1} s^{-1}	**Temp range/K**	**Ref**
6.94×10^{-13} exp(-496/T)	**211-374**	**Baasandorj et al[22]**
1.72×10^{-13}	296	Østerstrøm et al.[23]
4.80×10^{-13} exp(-445/T)	**253-328**	**Qing et al.[24]**

Rate constant (O_3) / cm^3 molecule^{-1} s^{-1}	**Temp range/K**	**Ref**
4.14×10^{-22}	**296**	**Østerstrøm et al.[23]**
$<5.20 \times 10^{-22}$	296	Baasandorj et al.[25]

Baasandorj et al.[22] and Qing et al.[24] agree from their temperature dependent studies that reaction obeys normal Arrhenius behaviour. Although the work of Baasandorj et al[22] covers a wider temperature range the work of Qing et al.[24] consistently yields rate coefficients between 10-20% lower. Therefore, we have integrated the model with both kinetic parametrisations. The work of Østerstrøm et al.[23] is adopted for the rate coefficient with ozone, consistent with the upper limit of Baasandorj et al.[25]

HFO-1336mzzZ $CF_3CH=CHCF_3$

Rate constant (OH) / cm^3 molecule^{-1} s^{-1}	**Temp range/K**	**Ref**
5.73×10^{-19} T^2 exp(678/T)	**211-374**	**Baasandorj et al[25]**
4.21×10^{-13}	296	Østerstrøm et al.[23]

Rate constant (O_3) / cm^3 molecule^{-1} s^{-1}	**Temp range/K**	**Ref**
6.25×10^{-22}	**296**	**Østerstrøm et al.[23]**
$<6.00 \times 10^{-21}$	296	Baasandorj et al.[25]

Baasandorj et al[25] is the only temperature dependent study and displays Anti-Arrhenius behaviour, with the room temperature study of Østerstrøm et al.[23] The work of Østerstrøm et al.[23] is adopted for the rate coefficient with ozone, consistent with the upper limit of Baasandorj et al.[25]

HFO-1318myE $CF_3CF=CFCF_3$

Rate constant (OH) / cm^3 $molecule^{-1}$ s^{-1}	**Temp range/K**	**Ref**
4.34×10^{-13}	296-375	Baasandorj et al[26]
7.50×10^{-14} $(T/298)^{1.68}$exp(612/T)	**230-370**	**Orkin et al.[27]**

Baasandorj et al[26] have studied the reaction over a range of temperature and report no significant change. The work of Orkin et al.[27] is over a wider temperature range and has been adopted for this work.

HFO-1318myZ $CF_3CF=CFCF_3$

Rate constant (OH) / cm^3 $molecule^{-1}$ s^{-1}	**Temp range/K**	**Ref**
3.30×10^{-13}	296-375	Baasandorj et al[26]
2.99×10^{-14} $(T/298)^{2.61}$exp(760/T)	**230-370**	**Orkin et al.[27]**

Baasandorj et al[26] have studied the reaction over a range of temperature and report no significant change. The work of Orkin et al.[27] is over a wider temperature range and has been adopted for this work.

HFO-1243zf $CF_3CH=CH_2$

Rate constant (OH) / cm^3 $molecule^{-1}$ s^{-1}	**Temp range/K**	**Ref**
8.28×10^{-13}exp(183/T)	252-370	Orkin et al.[5]
7.65×10^{-13}exp (165/T)	**263-358**	**Gonzalez et al.[28]**
1.36×10^{-12}	296	Andersen et al.[29]

Rate constant (O_3) / cm^3 $molecule^{-1}$ s^{-1}	**Temp range/K**	**Ref**
3.01×10^{-19}	**296**	**Soto et al.[30]**

There are two temperature dependent studies[5,28] that both display Anti-Arrhenius behaviour. The work of Gonzalez et al.[28] is adopted yielding smaller rate coefficients across the temperature range. The rate constant for the reaction with ozone used is that of Soto et al.[30]

HFO-1438ezyE $(CF_3)_2CFCH=CHF$

Rate constant (OH) / cm^3 $molecule^{-1}$ s^{-1}	**Temp range/K**	**Ref**
$7.34 \times 10^{-19}T^2$ exp(-481/T)	**214-380**	**Papadimitriou & Burkholder[31]**

The only experimental study by Papadimitriou & Burkholder[33] has been adopted for this work.

HCFO-1233xf $CF_3CCl=CH_2$

Rate constant (OH) / cm^3 $molecule^{-1}$ s^{-1}	**Temp range/K**	**Ref**
2.39×10^{-12}	**298**	**Thomsen & Jorgensen[32]**
9.43×10^{-13} exp(98/T)	**200-400**	**Michelat et al.[33]**

Rate constant (O_3) / cm^3 $molecule^{-1}$ s^{-1}	Temp range/K	Ref
3.54×10^{-21}	**296**	**McGillen et al.[9]**

No experimental study exists that reports the 1233xf+OH rate co-efficient. However, a theoretical study by Thomsen and Jorgensen[32] and a temperature dependent structure-activity relationship study by Michelat et al.[33] have been reported. There is one study for the reaction with ozone.[9]

HFO–1216 CF_2=$CFCF_3$

Rate constant (OH) / cm^3 $molecule^{-1}$ s^{-1}	Temp range/K	Ref
9.96×10^{-13} exp(244/T)	293-489	Mcllroy et al.[34]
5.66×10^{-13} exp(407/T)	252-370	Orkin et al.[5]
8.74×10^{-13} exp(260/T)	**250-430**	**Tokuhashi et al.[35]**
2.4×10^{-12}	296	Mashino et al.[36]
2.6×10^{-12}	296	Acerboni et al.[37]
9.75×10^{-14} (T/298) $^{1.94}$ exp(922/T)	**230-480**	**Orkin et al.[27]**

Rate constant (O_3) / cm^3 $molecule^{-1}$ s^{-1}	Temp range/K	Ref
6.2×10^{-22}	**296**	**Acerboni et al.[37]**
$< 3.10 \times 10^{-21}$	296	Mashino et al.[36]

There are four temperature dependent studies,[5,27, 34,35] if we ignore[5] and for reasons given earlier and focus on the two with the widest temperature range[27,35] both describe Anti-Arrhenius behaviour and rather than select one we have run integrations using both determinations. The work of Acerboni et al.[37] is used for the reaction with ozone, consistent with the upper limit reported by Mashino et al.[36]

Section 2: Lifetime, GWP and TFA yields.

The oxidation of these HFOs will lead to either $CF_3C(O)F$, i.e. HFO-1234yf, HFO-1225ye and HFO-1438ezy(E), where the CF_3CF moiety is present or CF_3CHO, i.e. HFO-1234ze, HFO-1336ze and HFO-1243zf. It is generally accepted that $CF_3C(O)F$ leads to 100% TFA production,[38] whereas for CF_3CHO, it depends on whether the compound is removed by reaction with OH or via photolysis or via deposition and so an estimate of 2-30% yield of TFA has been proposed.[38] Given that the reaction between OH and these HFO alkenes will proceed via addition of the OH to the double bond, there is the possibility that rate constants will have a pressure dependence, but from the studies carried out it appears that all the HFOs considered are at the high pressure limit and therefore no pressure dependence is observed. However, it is also possible for these addition reactions to display Anti-Arrhenius behaviour, i.e. that the rate constant increases with decreasing temperature.[39,40] The GWP_{100} will depend on the lifetime of the compound the intensity of its infrared absorption spectrum and how much it overlaps with the outgoing infrared spectrum of the Earth.

Table S1. Estimated lifetimes for HFOs and TFA yields.

HFO	Formula	Lifetime due to OH loss	TFA yield	References
1234yf	$CF_3CF=CH_2$	11 days	100%	Nielsen et al[4]
		12 days		Orkin et al.[6]
		12 days		Papadimitriou et al.[7]
		11 days		Tokuhashi et al.[8]
1225ye(E)	$CF_3CF=CHF$	10 days	100%	Hurley et al.[10]
1225ye(Z)	$CF_3CF=CHF$	18 days	100%	Hurley et al.[10]
		10 days	100%	Papadimitriou et al.[7]
1233zd(E)	$CF_3CH=CHCl$	34 days	2-30%	Gierczak et al[13]
		42 days	2-30%	UNEP[41]
1233zd(Z)	$CF_3CH=CHCl$	11 days	2-30%	Gierczak et al.[13],
		13 days	2-30%	UNEP[41]
1234ze(E)	$CF_3CH=CHF$	15 days	2-30%	Antinolo et al.[17],
		19 days	2-30%	UNEP[41]
1234ze(Z)	$CF_3CH=CHF$	8 days	2-30%	Antinolo et al.[17]
		12 days		Nilsson et al.[21]
		12.3 days		Xu et al.[42]
		9.9 days	2-30%	UNEP[41]
1336mzz(E)	$CF_3CH=CHCF_3$	90 days	4-60%	Baasandorj et al.[22]
		67 days		Østerstrøm et al.[23]
		105 days		Qing et al.[24]
		122 days	4-60%	UNEP[41]
1336mzz(Z)	$CF_3CH=CHCF_3$	20 days	4-60%	Baasandorj et al.[25]
		27 days		Østerstrøm et al.[23]
		27 days	4-60%	UNEP[41]
1318my(E)	$CF_3CF=CFCF_3$	25 days	100%	Baasandorj et al.[26]
		21 days		Orkin et al.[27]
1318my(Z)	$CF_3CF=CFCF_3$	35 days	100%	Baasandorj et al.[26]
		34 days		Orkin et al.[27]
1243zf	$CF_3CH=CH_2$	6 days	2-30%	Gonzalez et al.[29]
1438ezy(E)	$(CF_3)_2CFCH=CHF$	36 days	90%	Papadimitriou and Burkholder[31]
1233xf	$CF_3CCl=CH_2$		2-30%*	
1216	$CF_3CF=CF_2$	5.3 days	100%	Orkin et al.[27]

*the TFA yield value from the oxidation of HFO-1233xf is assumed to be same as HFO-1243zf

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387 **Table S2.** Details of each HFO oxidation simulation

Simulation	HFO	Emissions (Gg yr^{-1})	TFA yield	Global/Regional Emission
STOCH-HFO-HY	All 15 HFOs	150 (Each 10)	High	Global
STOCH-HFO-LY	All 15 HFOs	150 (Each 10)	Low	Global
STOCH-HFO-HY-1	All 15 HFOs	15 (Each 1)	High	Global
STOCH-HFO-LY-1	All 15 HFOs	15 (Each 1)	Low	Global
STOCH-HFO-HY-100	All 15 HFOs	1500 (Each 100)	High	Global
STOCH-HFO-LY-100	All 15 HFOs	1500 (Each 100)	Low	Global
STOCH-HFO-1234yf	HFO-1234yf	10	n/a	Global
STOCH-HFO-1225yeZ	HFO-1225yeZ	10	n/a	Global
STOCH-HFO-1225yeE	HFO-1225yeE	10	n/a	Global
STOCH-HCFO-1233zdZ-HY	HCFO-1233zdZ	10	High	Global
STOCH-HCFO-1233zdE-HY	HCFO-1233zdE	10	High	Global
STOCH-HFO-1234zeZ-HY	HFO-1234zeZ	10	High	Global
STOCH-HFO-1234zeE-HY	HFO-1234zeE	10	High	Global
STOCH-HFO-1336mzzmZ-HY	HFO-1336mzzmZ	10	High	Global
STOCH-HFO-1336mzzmE-HY	HFO-1336mzzmE	10	High	Global
STOCH-HFO-1318myZ	HFO-1318myZ	10	n/a	Global
STOCH-HFO-1318myE	HFO-1318myE	10	n/a	Global
STOCH-HFO-1243zf-HY	HFO-1243zf	10	High	Global
STOCH-HFO-1438ezyE	HFO-1438ezyE	10	n/a	Global
STOCH-HCFO-1233xf-HY	HCFO-1233xf	10	High	Global
STOCH-HFO-1216	HFO-1216	10	n/a	Global
STOCH-HCFO-1233zdZ-LY	HCFO-1233zdZ	10	Low	Global
STOCH-HCFO-1233zdE-LY	HCFO-1233zdE	10	Low	Global
STOCH-HFO-1234zeZ-LY	HFO-1234zeZ	10	Low	Global
STOCH-HFO-1234zeE-LY	HFO-1234zeE	10	Low	Global
STOCH-HFO-1336mzzmZ-LY	HFO-1336mzzmZ	10	Low	Global
STOCH-HFO-1336mzzmE-LY	HFO-1336mzzmE	10	Low	Global
STOCH-HFO-1243zf-LY	HFO-1243zf	10	Low	Global

STOCH-HCFO-1233xf-LY	HCFO-1233xf	10	Low	Global
STOCH-HFO1234yf-AS	HFO1234yf	2.7	n/a	Asia
STOCH-HFO1234yf-EU	HFO1234yf	4.0	n/a	Europe
STOCH-HFO1234yf-NA	HFO1234yf	2.7	n/a	North America
STOCH-HFO1216-AS	HFO1216	2.7	n/a	Asia
STOCH-HFO1216-EU	HFO1216	4.0	n/a	Europe
STOCH-HFO1216-NA	HFO1216	2.7	n/a	North America
STOCH-HFO1336mzzE-HY-AS	HFO1336mzzmE	2.7	High	Asia
STOCH-HFO1336mzzE-HY-EU	HFO1336mzzmE	4.0	High	Europe
STOCH-HFO1336mzzE- HY-NA	HFO1336mzzmE	2.7	High	North America

n/r represents not available data

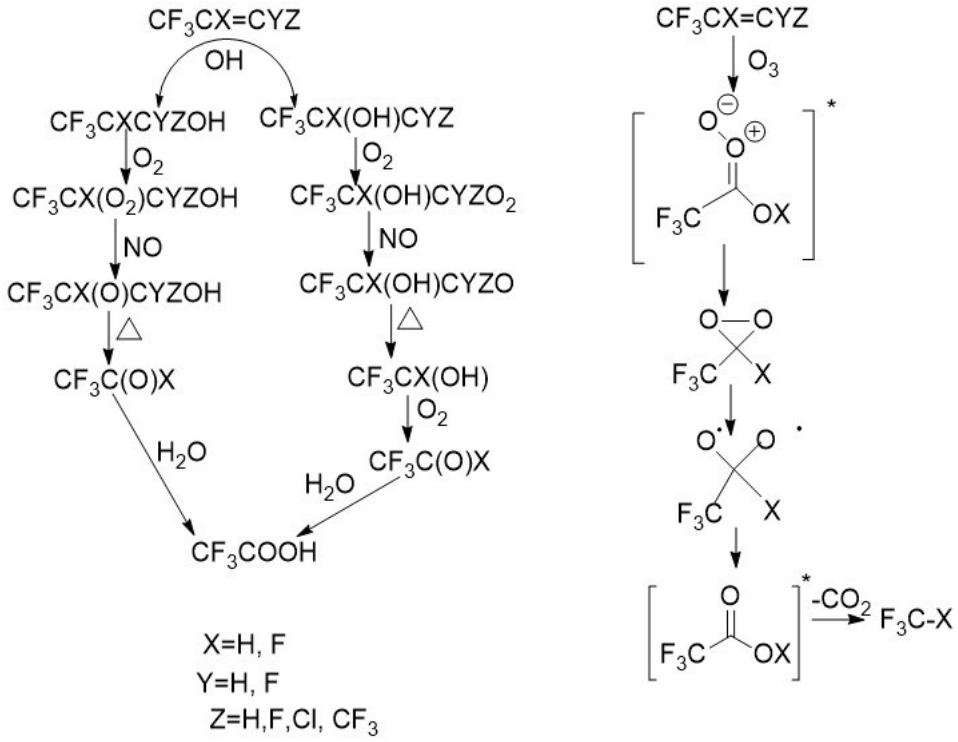


Figure S1. The oxidation of HFO by OH (left) and O_3 (right) producing TFA

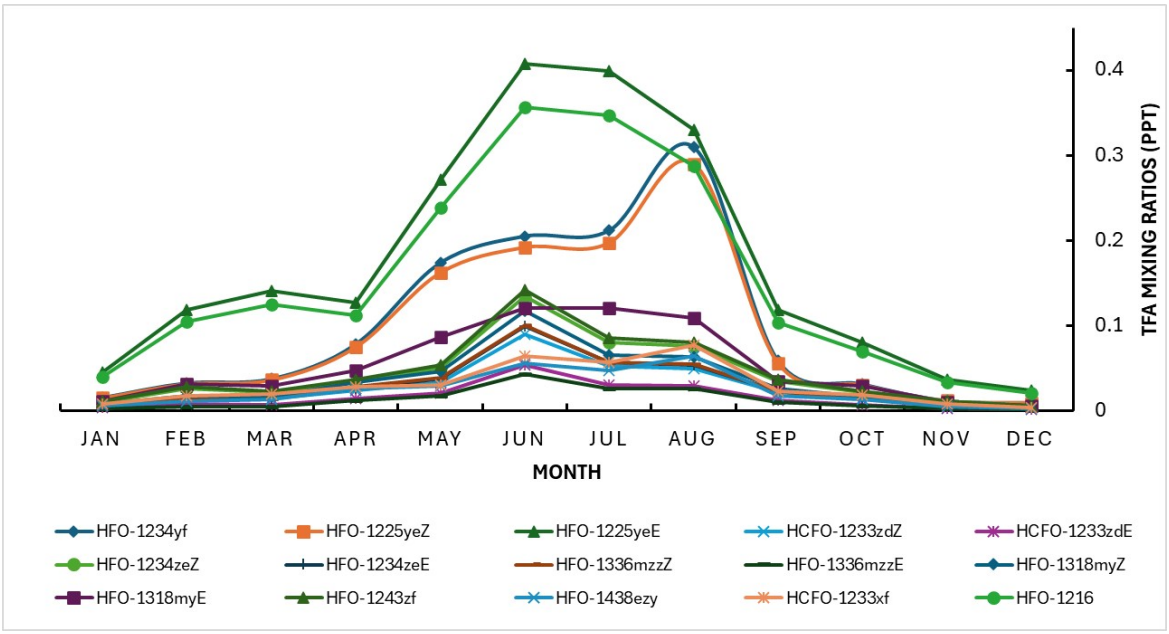


Figure S2. Monthly variation of maximum TFA mixing ratios (fixed grid cell) produced from the individual HFO oxidation reactions in the model

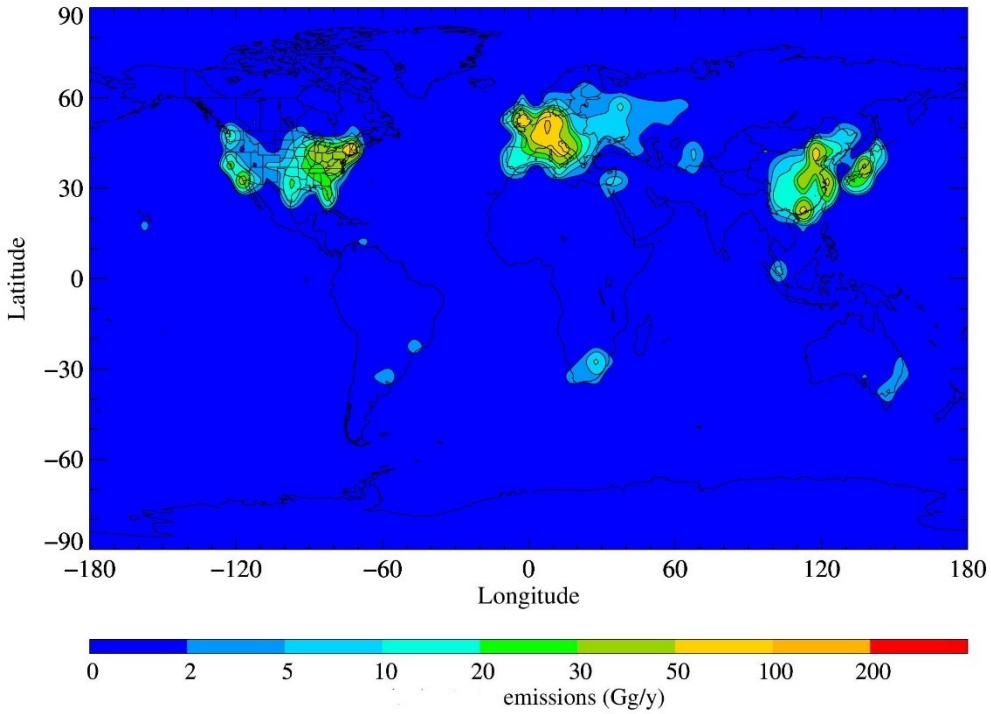


Figure S3. Annual HFO emission distribution adapted from 2010 HFC-134a EDGAR emission inventory

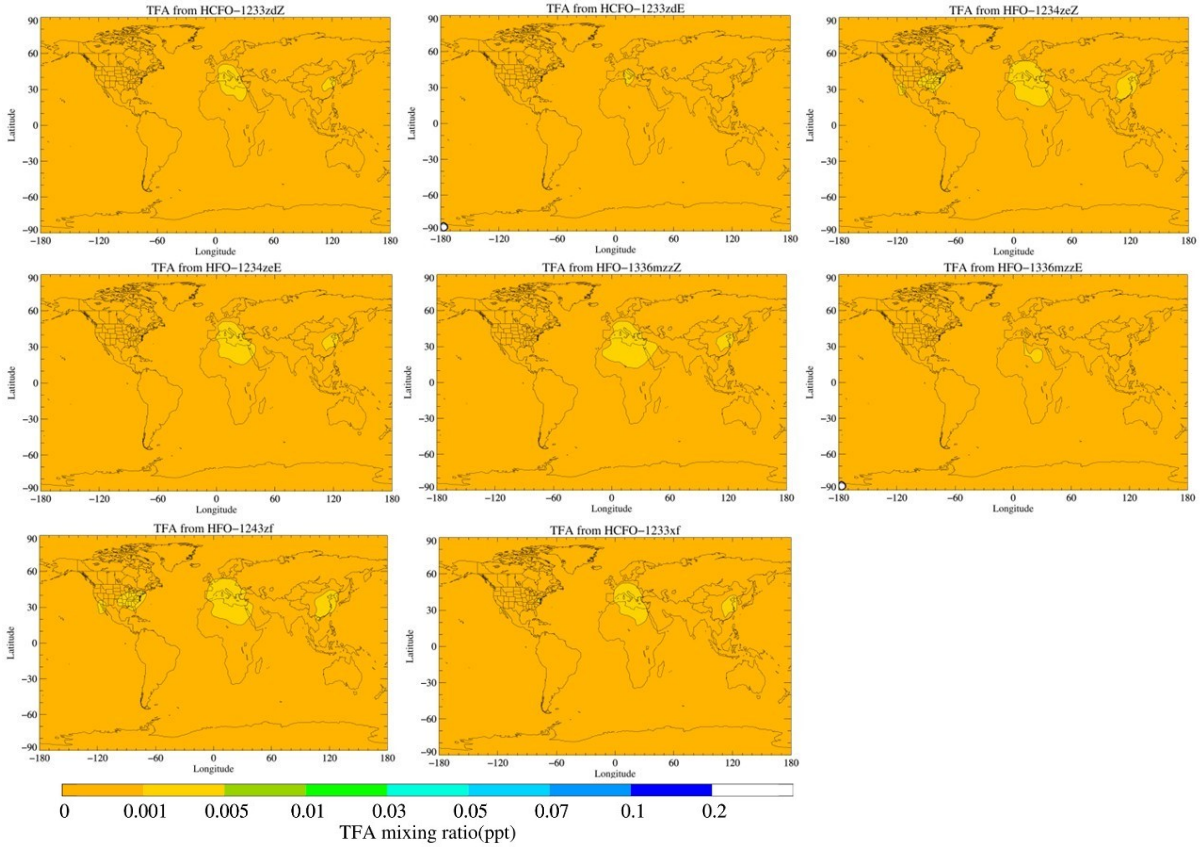
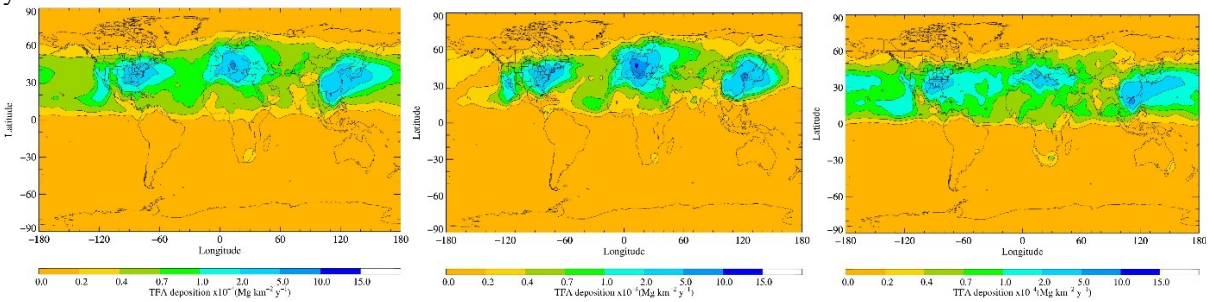
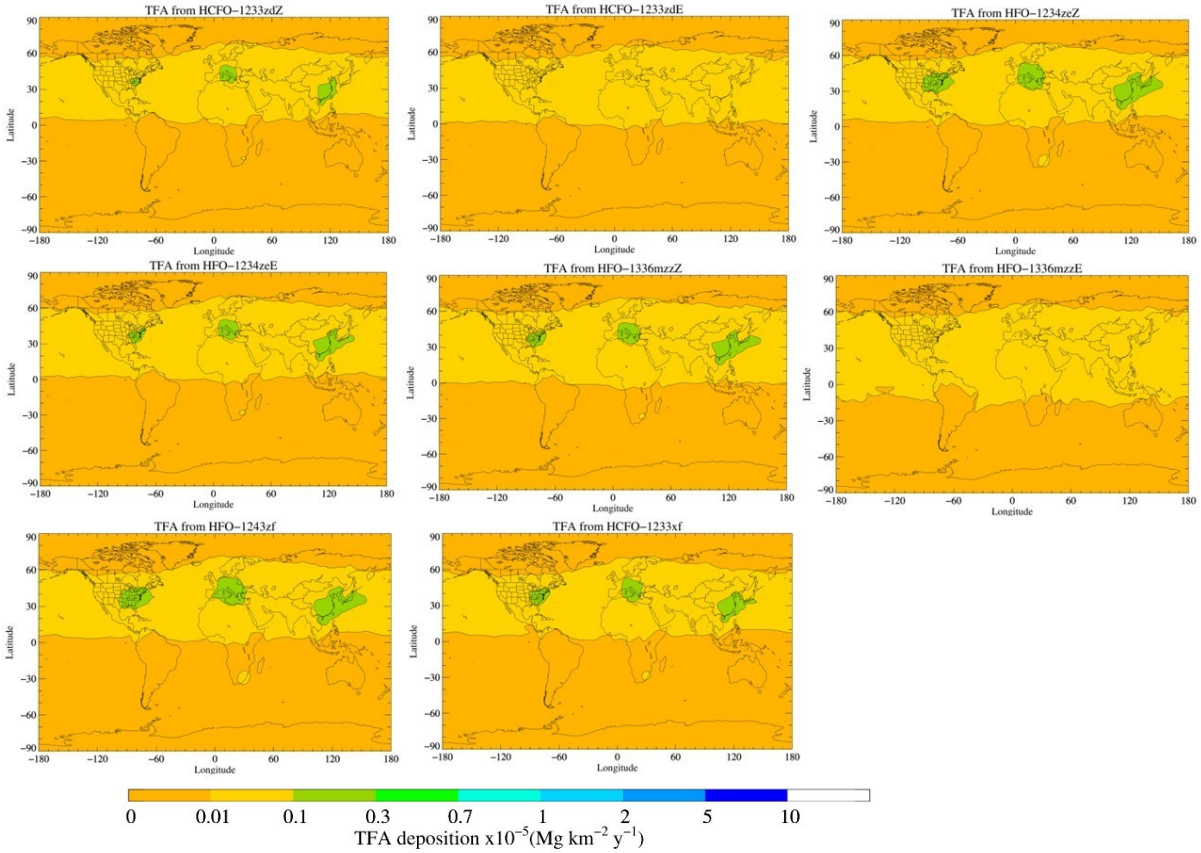


Figure S4. Annual surface distribution plots of TFA mixing ratios produced from eight HFOs with lower TFA yields



(a) (b) (c)

Figure S5. Surface distribution plots of TFA deposition simulated by the STOCH-HFO-LY, (a) Annual, (b) June-July-August, (c) December-January-February



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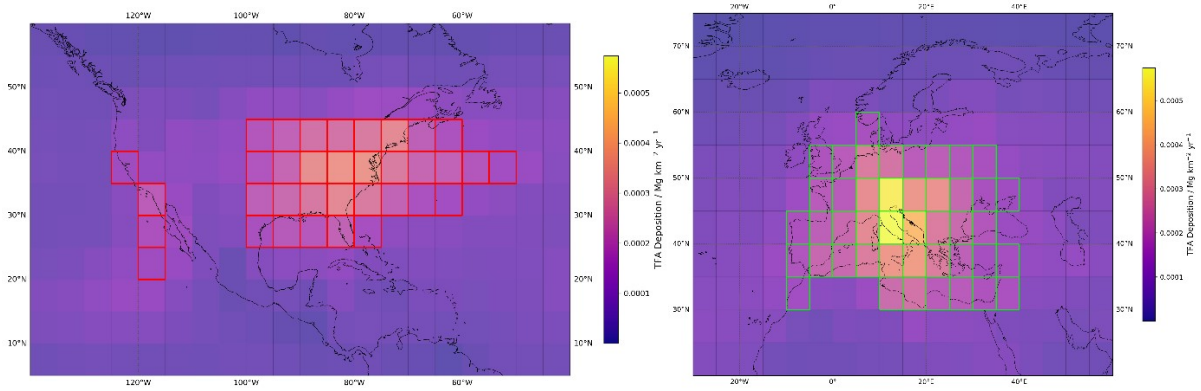
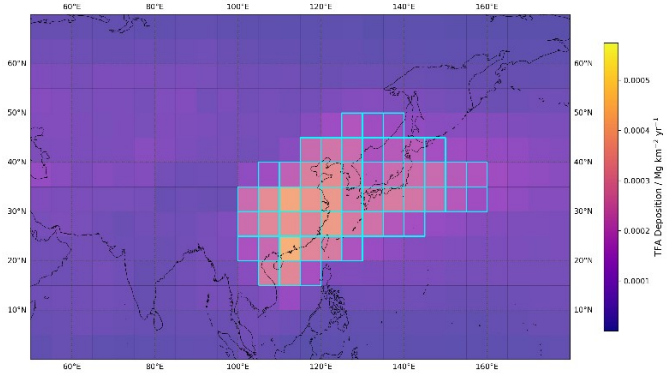
411

412 **Figure S6.** Annual surface distribution plots of TFA depositions produced from eight HFOs with lower TFA
413 yields

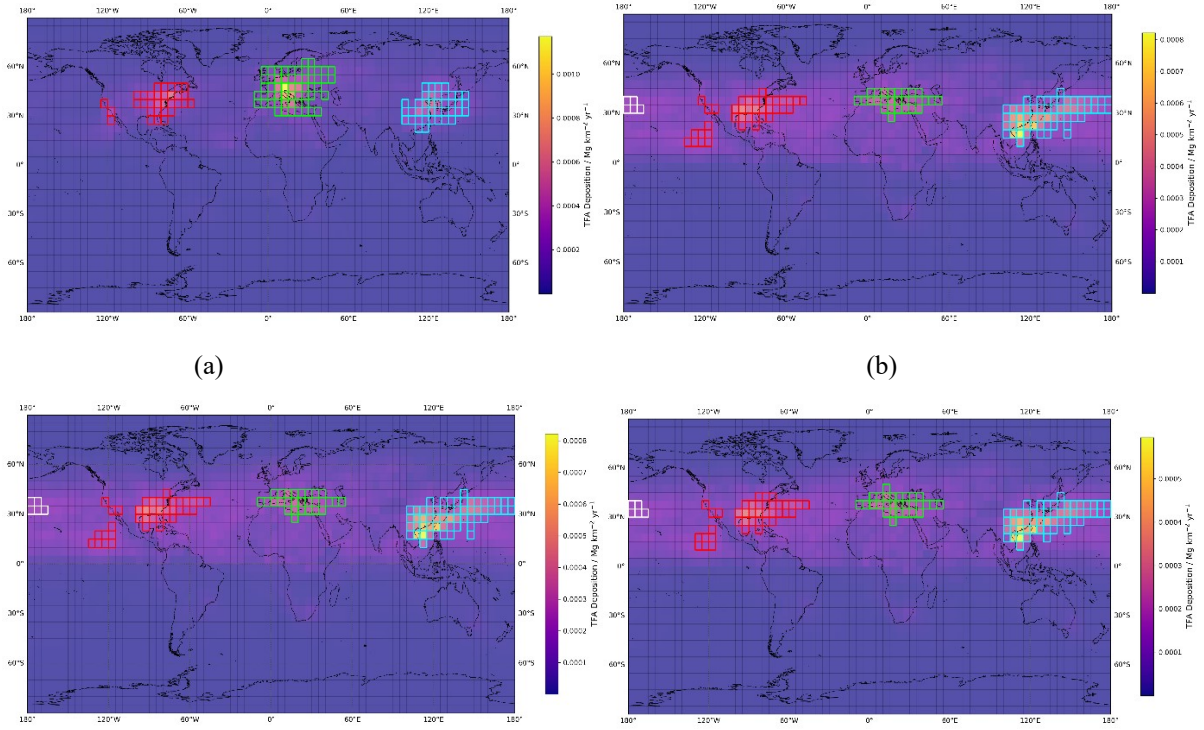


414
415 (a) (b)
416



417
418 (c)

419 **Figure S7.** The hot spots for TFA deposition flux per grid box simulated by the STOCH-HFO-LY model (a)
420 North America (b) Europe, and (c) Asia



(a) (b)

(c) (d)

Figure S8. The hot spots of TFA deposition flux during June-July-August (a) December-January-February (b) simulated by the STOCH-HFO-HY, the hot spots of TFA deposition flux during June-July-August (c) December-January-February (d) simulated by the STOCH-HFO-LY

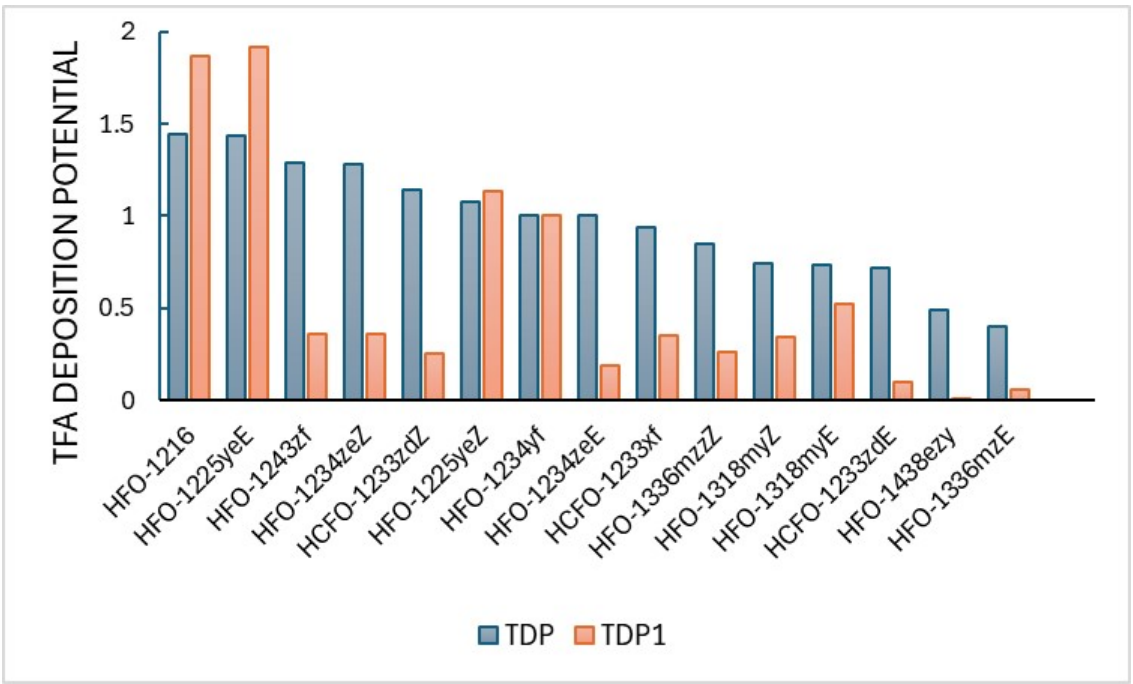


Figure S9. Comparative TFA deposition potential from the model study (TDP) and simple kinetic study (TDP1). TDP and TDP1 calculations from the lower limit of TFA yields have not been included in the figure

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