

# IUPAC Task Group on Atmospheric Chemical Kinetic Data Evaluation – Data Sheet HO<sub>x</sub>\_VOC8

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This data sheet last evaluated June 2013 (with no revision of the preferred values).

## HO + CH<sub>2</sub>=C(CH<sub>3</sub>)CH=CH<sub>2</sub> (isoprene) → products

### Rate coefficient data

$k/\text{cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$	Temp./K	Reference	Technique/ Comments
<i>Absolute Rate Coefficients</i>			
$2.36 \times 10^{-11} \exp[(409 \pm 28)/T]$	299-422	Kleindienst et al., 1982	FP-RF
$(9.26 \pm 1.5) \times 10^{-11}$	299		
$9.7 \times 10^{-11} (T/298)^{-1.36}$	249-348	Siese et al., 1994	FP-RF
$9.7 \times 10^{-11}$	298		
$(1.10 \pm 0.05) \times 10^{-10}$	298	Stevens et al., 1999	DF-LIF
$2.70 \times 10^{-11} \exp[(336 \pm 74)/T]$	251-342	Campuzano-Jost et al., 2000	PLP-LIF (a)
$(8.56 \pm 0.26) \times 10^{-11}$	297		
$(1.10 \pm 0.04) \times 10^{-10}$	300	Chuong and Stevens, 2000	DF-LIF (b)
$(9.6 \pm 0.5) \times 10^{-11}$	295	McGivern et al., 2000	PLP-LIF (c)
$(1.01 \pm 0.08) \times 10^{-10}$	298 ± 2	Zhang et al., 2000	DF-CIMS (d)
$9.1 \times 10^{-11}$	298 ± 2	Zhang et al., 2001	DF-CIMS (e)
$(1.08 \pm 0.05) \times 10^{-10}$	300	Chuong and Stevens, 2002	DF-LIF (f)
$2.68 \times 10^{-11} \exp[(348 \pm 136)/T]$	251-342	Campuzano-Jost et al., 2004	PLP-LIF (g)
$(8.47 \pm 0.59) \times 10^{-11}$	297		
$(1.00 \pm 0.15) \times 10^{-10}$	293	Spangenberg et al., 2004	PLP-LIF (h)
$(1.00 \pm 0.12) \times 10^{-10}$	294 ± 1.1	Karl et al., 2004	(i)
$(1.02 \pm 0.09) \times 10^{-10}$	295 ± 1	Poppe et al., 2007	(i)
$(1.12 \pm 0.07) \times 10^{-10}$	296 ± 2		
$(1.07 \pm 0.08) \times 10^{-10}$	290.5 ± 2.5		
$(9.7 \pm 0.8) \times 10^{-11}$	291 ± 2		
$(9.9 \pm 0.8) \times 10^{-11}$	290 ± 1		
<i>Relative Rate Coefficients</i>			
$7.8 \times 10^{-11}$	300	Cox et al., 1980	RR (j)
$(9.98 \pm 0.45) \times 10^{-11}$	299 ± 2	Atkinson et al., 1982	RR (k)
$(9.90 \pm 0.27) \times 10^{-11}$	297 ± 2	Ohta, 1983	RR (l)
$(1.02 \pm 0.04) \times 10^{-10}$	295 ± 1	Atkinson and Aschmann, 1984	RR (m)
$(1.01 \pm 0.02) \times 10^{-10}$	297 ± 1	Edney et al., 1986	RR (n)
$(1.11 \pm 0.23) \times 10^{-10}$	298	McQuaid et al., 2002	RR (o)
$2.54 \times 10^{-11} \exp[(409 \pm 42)/T]$	298-363	Gill and Hites, 2002	RR (p)
$(1.00 \pm 0.05) \times 10^{-10}$	298		
$(1.03 \pm 0.04) \times 10^{-10}$	298 ± 2	Iida et al., 2002	RR (q)
$2.33 \times 10^{-11} \exp[(444 \pm 27)/T]$	240-340	Singh and Li, 2007	RR (r)
$(1.07 \pm 0.03) \times 10^{-10}$	298	Singh and Li, 2007	RR (r,s)
$(1.11 \pm 0.02) \times 10^{-10}$	298	Singh and Li, 2007	RR (r,t)
$3.97 \times 10^{-11} \exp[(249 \pm 20)/T]$	323-413	Hites and Turner, 2009	RR (p)

## Comments

- (a) Rate coefficients for the reactions of HO radicals with isoprene-d<sub>6</sub> and DO radicals with isoprene were also measured at 297 K, these being  $(8.31 \pm 0.10) \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  and  $(8.27 \pm 0.10) \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ , respectively.
- (b) Carried out at total pressures of 2-6 Torr (2.7-8.0 mbar) of helium diluent. No pressure dependence of the rate coefficient was observed at 300 K. However, at higher temperatures (321-423 K) the rate coefficient increased with increasing total pressure, with the effect being more pronounced at higher temperature.
- (c) Carried out at total pressures of 0.5-20 Torr (0.7-27 mbar) of argon diluent. The rate coefficient was observed to be pressure dependent below 10 Torr total pressure; the value cited in the table is that measured at 20 Torr total pressure.
- (d) Carried out at total pressures of 72.7-112.7 Torr (97-150 mbar) of N<sub>2</sub> diluent. The measured rate coefficient was independent of pressure over this range.
- (e) Based on a very limited kinetic study carried out at a total pressure of 1.9 Torr (2.5 mbar) of helium diluent (the focus of the study was on formation of HO-isoprene adducts and their subsequent reactions).
- (f) Carried out at total pressures of argon diluent of 100 Torr (133 mbar) and 150 Torr (200 mbar). The measured rate coefficient was independent of total pressure over this range.
- (g) Rate coefficients for HO + isoprene-d<sub>6</sub> and DO + isoprene were also measured at 297 K, with values of  $(8.27 \pm 0.17) \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  and  $(8.43 \pm 0.18) \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ , respectively. Isoprene and isoprene-d<sub>6</sub> concentrations in the gas flow stream were measured before and after the reaction cell by UV absorption at 228.9 nm. The data from this study are in excellent agreement with those previously reported by Campuzano-Jost et al. (2000) at the same temperatures, and are assumed to supersede the earlier study.
- (h) Rate coefficients were also measured at 58, 71, 84, 104 and 114 K, with the rate coefficients of  $(7.8 \pm 1.2) \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ ,  $(1.14 \pm 0.17) \times 10^{-10} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ ,  $(9.8 \pm 1.5) \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ ,  $(1.88 \pm 0.28) \times 10^{-10} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  and  $(1.68 \pm 0.25) \times 10^{-10} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ , respectively.
- (i) Carried out in a large volume (270 m<sup>3</sup>) chamber. HO radicals were measured by LIF and isoprene was measured by GC (Karl et al., 2004; Poppe et al., 2007) or by proton-transfer-reaction mass spectrometry (Poppe et al., 2007) during irradiations of air mixtures containing low concentrations of isoprene and NO<sub>x</sub>.
- (j) Relative rate method carried out at atmospheric pressure of air. HO radicals were generated by photolysis of HONO at wavelengths >300 nm. The concentrations of isoprene and ethene (the reference compound) were measured by GC. The measured rate coefficient ratio  $k(\text{HO} + \text{isoprene})/k(\text{HO} + \text{ethene})$  is placed on an absolute basis by use of a rate coefficient of  $k(\text{HO} + \text{ethene}) = 8.44 \times 10^{-12} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  at 300 K and atmospheric pressure of air (Atkinson and Arey, 2003).
- (k) Relative rate method carried out at atmospheric pressure of air. HO radicals were generated by photolysis of CH<sub>3</sub>ONO at wavelengths >300 nm. The concentrations of isoprene and propene (the reference compound) were measured by GC. The measured rate coefficient ratio of  $k(\text{HO} + \text{isoprene})/k(\text{HO} + \text{propene}) = 3.81 \pm 0.17$  is placed on an absolute basis by use of a rate coefficient of  $k(\text{HO} + \text{propene}) = 2.62 \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  at 299 K and atmospheric pressure of air (Atkinson and Arey, 2003).
- (l) Relative rate method carried out at atmospheric pressure of N<sub>2</sub> + O<sub>2</sub>. HO radicals were generated by photolysis of H<sub>2</sub>O<sub>2</sub> at 253.7 nm. The concentrations of isoprene and 1,3-butadiene (the reference compound) were measured by GC. The measured rate coefficient

ratio of  $k(\text{HO} + \text{isoprene})/k(\text{HO} + 1,3\text{-butadiene}) = 1.48 \pm 0.04$  is placed on an absolute basis by use of a rate coefficient of  $k(\text{HO} + 1,3\text{-butadiene}) = 6.69 \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  at 297 K and atmospheric pressure of air (Atkinson and Arey, 2003).

- (m) Relative rate method carried out at atmospheric pressure of air. HO radicals were generated by photolysis of  $\text{CH}_3\text{ONO}$  at wavelengths  $>300 \text{ nm}$ . The concentrations of isoprene and propene (the reference compound) were measured by GC. The measured rate coefficient ratio of  $k(\text{HO} + \text{isoprene})/k(\text{HO} + \text{propene}) = 3.81 \pm 0.15$  is placed on an absolute basis by use of a rate coefficient of  $k(\text{HO} + \text{propene}) = 2.68 \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  at 295 K and atmospheric pressure of air (Atkinson and Arey, 2003).
- (n) Relative rate method carried out at atmospheric pressure of air. HO radicals were generated by photolysis of  $\text{CH}_3\text{ONO}$  at wavelengths  $>300 \text{ nm}$ . The concentrations of isoprene and propene (the reference compound) were measured by GC. The measured rate coefficient ratio of  $k(\text{HO} + \text{isoprene})/k(\text{HO} + \text{propene})$  is placed on an absolute basis by use of a rate coefficient of  $k(\text{HO} + \text{propene}) = 2.65 \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  at 297 K and atmospheric pressure of air (Atkinson and Arey, 2003).
- (o) Relative rate method carried out at atmospheric pressure of air. HO radicals were generated by photolysis of  $\text{CH}_3\text{ONO}$ . The concentrations of isoprene and *trans*-2-butene (the reference compound) were measured by GC. The measured rate coefficient ratio of  $k(\text{HO} + \text{isoprene})/k(\text{HO} + \textit{trans}\text{-2-butene}) = 1.74 \pm 0.14$  is placed on an absolute basis by use of a rate coefficient of  $k(\text{HO} + \textit{trans}\text{-2-butene}) = 6.40 \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  at 298 K and atmospheric pressure of air (IUPAC, current recommendation). The cited error limits are the 95% confidence intervals (McQuaid et al., 2002).
- (p) Relative rate method carried out in a  $192 \text{ cm}^3$  volume quartz reaction vessel at atmospheric pressure of helium diluent. HO radicals were generated by photolysis of  $\text{H}_2\text{O}_2$ . The concentrations of isoprene and 2-methylpropene (the reference compound) were measured by MS. Rate coefficients were measured over the temperature ranges 298–363 K by Gill and Hites (2002) and 323–413 K by Hites and Turner (2009). The measured rate coefficient ratios of  $k(\text{HO} + \text{isoprene})/k(\text{HO} + 2\text{-methylpropene})$  are placed on an absolute basis by use of a rate coefficient of  $k(\text{HO} + 2\text{-methylpropene}) = 9.4 \times 10^{-12} \exp(505/T) \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  (IUPAC, current recommendation). The cited Arrhenius expression for the Hites and Turner (2009) study is an un-weighted least-squares fit to their data; the cited error in the temperature dependence is two standard deviations. Combination of the rate coefficients of Gill and Hites (2002) and Hites and Turner (2009) results in the Arrhenius expression  $k = 3.12 \times 10^{-11} \exp[(339 \pm 19)/T] \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  or, because the combined data suggested a curved Arrhenius plot,  $k = 3.44 \times 10^{-17} T^2 \exp[(1037 \pm 14)/T] \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  (Hites and Turner, 2009), both relative to  $k(\text{HO} + 2\text{-methylpropene}) = 9.4 \times 10^{-12} \exp(505/T) \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  (IUPAC, current recommendation).
- (q) Carried out at atmospheric pressure of air. HO radicals were generated by photolysis of  $\text{CH}_3\text{ONO}$ . The concentrations of isoprene and cyclohexane, di-*n*-butyl ether or propene (the reference compounds) were measured by FTIR spectroscopy. The measured rate coefficient ratios of  $k(\text{HO} + \text{isoprene})/k(\text{HO} + \text{cyclohexane}) = 14.3 \pm 1.2$  and  $1.49 \pm 1.3$ ,  $k(\text{HO} + \text{isoprene})/k(\text{HO} + \text{di-}n\text{-butyl ether}) = 3.71 \pm 0.14$  and  $3.61 \pm 0.23$ , and  $k(\text{HO} + \text{isoprene})/k(\text{HO} + \text{propene}) = 3.89 \pm 0.20$ ,  $3.96 \pm 0.18$ ,  $3.90 \pm 0.15$  and  $3.98 \pm 0.20$  are placed on an absolute basis by use of rate coefficients at 298 K and atmospheric pressure of air of  $k(\text{HO} + \text{cyclohexane}) = 6.97 \times 10^{-12} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  (Atkinson and Arey, 2003),  $k(\text{HO} + \text{di-}n\text{-butyl ether}) = 2.78 \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  (Mellouki et al., 1995) and  $k(\text{HO} + \text{propene}) = 2.63 \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  (Atkinson and Arey, 2003). The value in the table is the un-weighted average together with the two standard deviation error.

- (r) Discharge flow system with MS detection of isoprene and the reference compounds dimethyl disulfide and ethanethiol. HO radicals generated by the reaction  $F + H_2O$ . Total pressure was in the range 1-3 Torr (1.3-4 mbar). The rate coefficient for the HO + isoprene reaction was independent of pressure over the range 1-3 Torr at 298 K, but was increased with increasing pressure over the range 1-3 Torr at 340 K. Temperature-dependent rate measurements were carried out at 1.0-1.1 Torr pressure, and hence the rate coefficient at 340 K would have been in the fall-off region. The measured rate coefficient ratios (which were tabulated only at 298 K) were placed on an absolute basis using  $k(\text{HO} + \text{dimethyl disulfide}) = 5.9 \times 10^{-11} \exp(380/T) \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  (Wine et al., 1981) and  $k(\text{HO} + \text{ethanethiol}) = 1.23 \times 10^{-11} \exp(396/T) \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  (Wine et al., 1984). The rate coefficient used for  $k(\text{HO} + \text{dimethyl disulfide})$  is slightly different from the current IUPAC recommendation of  $7.0 \times 10^{-11} \exp(350/T) \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  (IUPAC, 2008), but re-evaluation is only possible for the 298 K rate coefficient (see Comment (s) below).
- (s) Relative to dimethyl disulfide. The measured rate coefficient ratio  $k(\text{HO} + \text{isoprene})/k(\text{HO} + \text{dimethyl disulfide}) = 0.464 \pm 0.010$  at 298 K is placed on an absolute basis using  $k(\text{HO} + \text{dimethyl disulfide}) = 2.3 \times 10^{-10} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  (IUPAC, current recommendation).
- (t) Relative to ethanethiol. The measured rate coefficient ratio  $k(\text{HO} + \text{isoprene})/k(\text{HO} + \text{ethanethiol}) = 2.398 \pm 0.043$  at 298 K is placed on an absolute basis using  $k(\text{HO} + \text{ethanethiol}) = 4.64 \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  (Wine et al., 1984).

### Preferred Values

$k = 1.0 \times 10^{-10} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  at 298 K.

$k = 2.7 \times 10^{-11} \exp(390/T) \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  over the temperature range 240-430 K.

### Reliability

$\Delta \log k = \pm 0.06$  at 298 K.

$\Delta(E/R) = \pm 100 \text{ K}$ .

### Comments on Preferred Values

The room temperature absolute and relative rate coefficients of Kleindienst et al. (1982), Atkinson et al. (1982), Ohta (1983), Atkinson and Aschmann (1984), Edney et al. (1986), Siese et al. (1994), Stevens et al. (1999), Chuong and Stevens (2000, 2002), McGivern et al. (2000), Zhang et al. (2000), McQuaid et al. (2002), Iida et al. (2002), Gill and Hites (2002), Spangenberg et al. (2004), Karl et al. (2004), Poppe et al. (2007) and Singh and Li (2007) are in excellent agreement, with the absolute rate coefficients of Campuzano-Jost et al. (2000, 2004) being ~15% lower. The temperature dependencies obtained by Kleindienst et al. (1982), Siese et al. (1994), Campuzano-Jost et al. (2000, 2004), Gill and Hites (2002) and Singh and Li (2007) are also in good agreement (the temperature dependence measured by Siese et al. (1994) corresponds to  $E/R = -400 \text{ K}$  over the temperature range 249-349 K). The preferred temperature dependence is an average of those obtained from the temperature dependent studies of Kleindienst et al. (1982), Siese et al. (1994), Campuzano-Jost et al. (2000) and Gill and Hites (2002). The preferred 298 K rate coefficient is based on the room temperature rate coefficients of Kleindienst et al. (1982), Atkinson et al. (1982), Ohta (1983), Atkinson and Aschmann (1984), Edney et al. (1986), Siese et al. (1994), Stevens et al. (1999), Chuong and Stevens (2000, 2002), McGivern et al. (2000), Zhang et al. (2000), McQuaid et al. (2002), Iida et al. (2002), Gill and Hites (2002), Campuzano-Jost et al. (2004), Spangenberg et al. (2004), Karl et al. (2004) and Poppe et al. (2007), corrected to 298 K where necessary

using the preferred temperature dependence. The pre-exponential factor is calculated from the preferred 298 K rate coefficient and the preferred temperature dependence.

The reaction proceeds by initial addition of the HO radical to the two C=C bonds, and theoretical calculations predict that the percentages of HO radical addition to the 1-, 2-, 3- and 4-position carbon atoms in isoprene (2-methyl-1,3-butadiene) are 67%, 2%, 2% and 29%, respectively, at 300 K (Greenwald et al., 2007). Addition of O<sub>2</sub> then leads to the formation of six possible hydroxyalkyl radicals (Atkinson, 1997). The subsequent chemistry of these radicals in the atmosphere and the products observed is discussed by Atkinson (1997) and Calvert et al. (2000). At atmospheric pressure in the presence of NO (so that organic peroxy radicals react dominantly with NO) the products observed (Atkinson, 1997; Atkinson et al., 1989; Tuazon and Atkinson, 1990; Paulson et al., 1992; Miyoshi et al., 1994; Kwok et al., 1995; Chen et al., 1998; Sprengnether et al., 2002; Zhao et al., 2004; Baker et al., 2005; Karl et al., 2006) are methyl vinyl ketone (32-44%), methacrolein (22-28%), formaldehyde (57-66%), 3-methylfuran (≤5%), hydroxynitrates such as HOCH<sub>2</sub>C(CH<sub>3</sub>)=CHCH<sub>2</sub>ONO<sub>2</sub> (and isomers) (4-13%), and the hydroxycarbonyls HOCH<sub>2</sub>C(CH<sub>3</sub>)=CHCHO and HOCH<sub>2</sub>CH=C(CH<sub>3</sub>)CHO. 3-Methylfuran is almost certainly formed from cyclization and dehydration of HOCH<sub>2</sub>C(CH<sub>3</sub>)=CHCHO and HOCH<sub>2</sub>CH=C(CH<sub>3</sub>)CHO.

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- ▽ Karl et al. (2004); Poppe et al. (2007)
- Atkinson et al. (1982); Atkinson and Aschmann (1984)
- ◆ Ohta (1983)
- ▲ Edney et al. (1986)
- ◆ McQuaid et al. (2002)
- Gill and Hites (2002)
- ▼ Iida et al. (2002)
- Hites and Turner (2009)
- Singh and Li (2007)

