

Supplementary Information

S1. Laboratory characterizations of NO, NO₂ and ozone interferences

S1.1 NO interference

To quantify the NO interference, NO from a standard gas cylinder (Air Liquide, Krefeld, Germany) was added to the CRM system running in C2 mode at pyrrole/OH ≈ 2.0. The mixing ratios of the added NO were ~0, 5, 10, 15, 20, 30, 90, 140, 170, 220, 350 ppb. The resulting decrease in pyrrole mixing ratio (ΔC_3 in ppb) vs. the NO mixing ratio resulted in a polynomial relationship of the form $\Delta C_3 = a[NO]^2 + b[NO]$. During AQABA: $a \approx -3E-4$; $b \approx 0.17$.

S1.2 NO₂ interference

To quantify the NO₂ interference, NO₂ from a standard gas cylinder (Westfalen AG, Münster, Germany) was added to the CRM system running in C2 mode at pyrrole/OH ≈ 2.0. All tubes were flushed with NO₂ for several hours and the contact of the NO₂ with metal surfaces (pressure reducer, mass flow controller) was reduced to a minimum to avoid losses. To quantify the amount of NO₂ lost/converted to NO in the setup used, conversion to NO was monitored by an NO/NO₂ analyzer (Two-channel-chemiluminescence detector). 8–12 % of the NO₂ was converted to NO depending on the flows and mixing ratios used. This loss was corrected for when calculating the NO₂ mixing ratios in the reactor. The mixing ratios of the added NO₂ were ~0, 20, 40, 60, 100 ppb. The resulting decrease in pyrrole mixing ratio (ΔC_3 in ppb) vs. the NO₂ mixing ratio resulted in a polynomial relationship of the form $\Delta C_3 = a[NO_2]^2 + b[NO_2]$. During AQABA: $a \approx -2E-4$; $b \approx 0.08$.

S1.3 Ozone interference

Using an ozone generator (Thermo 49C O₃ Calibrator, Thermo Environmenal Instruments LLC, Franklin, USA), ozone was added to the CRM system running in C2 mode at pyrrole/OH ≈ 2.0. Ozone mixing ratios used were ~0, 20, 30, 45, 90, 130, 180, 260, 350 ppb. The resulting decrease in pyrrole mixing ratio (ΔC_3 in ppb) vs. the O₃ mixing ratio resulted in a linear relationship of the form $\Delta C_3 = a[O_3]$. During AQABA: $a \approx 0.0058$.

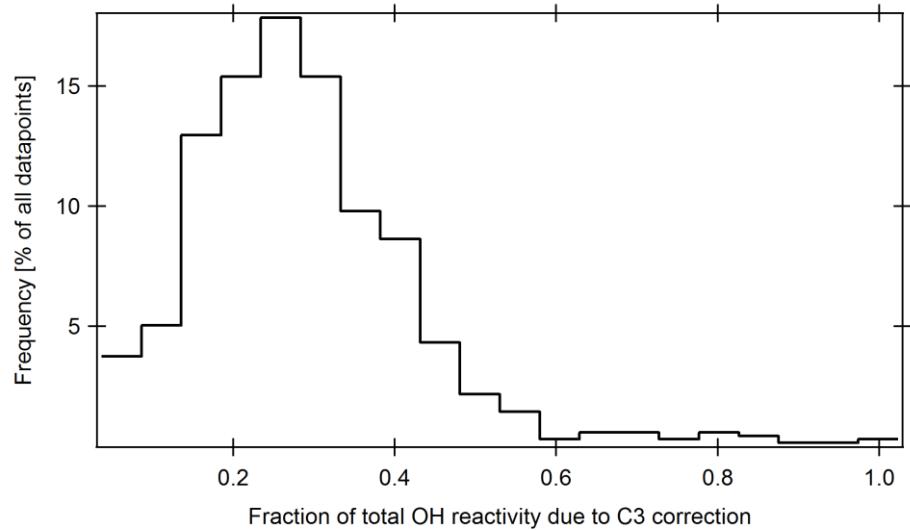


Figure S1. Frequency distribution of total OH reactivity fractions due to the NO, NO₂ and ozone corrections (C3 correction) for all datapoints in 50 min time resolution.

Table S1: Chemical species used for calculating speciated OH reactivity

Compound/ protonated mass (grey shaded compounds are gas standard calibrated with low uncertainty)	Chemical formula	k_{M+OH} [cm ³ molecules ⁻¹ s ⁻¹]	Instrument	Compounds attributed to protonated mass	If no unambiguous compound identification: reaction rate coefficient derived from...	Literature reference for reaction rate coefficient	Max. OH reactivity during AQABA [s ⁻¹]	Comment
Alkanes								
Methane	CH ₄	6.45E-15	Picarro Cavity ring-down spectroscopy instrument			IUPAC preferred value	0.35	
Ethane	C ₂ H ₆	2.40E-13	GC-FID			IUPAC preferred value	0.26	
Propane	C ₃ H ₈	1.10E-12	GC-FID			IUPAC preferred value	1.33	
2-Methylpropane	C ₄ H ₁₀	2.12E-12	GC-FID			Atkinson and Arey, 2003	0.68	
n-Butane	C ₄ H ₁₀	2.35E-12	GC-FID			IUPAC preferred value	1.42	
2-Methylbutane	C ₅ H ₁₂	3.60E-12	GC-FID			Atkinson and Arey, 2003	0.92	
n-pentane	C ₅ H ₁₂	3.80E-12	GC-FID			Atkinson and Arey, 2003	1.03	
2-Methylpentane	C ₆ H ₁₄	5.20E-12	GC-FID			Atkinson and Arey, 2003	0.11	
n-hexane	C ₆ H ₁₄	5.20E-12	GC-FID			Atkinson and Arey, 2003	0.16	
2,2,4- Trimethylpentane	C ₈ H ₁₈	3.34E-12	GC-FID			Atkinson and Arey, 2003	0.004	
n-heptane	C ₇ H ₁₆	6.76E-12	GC-FID			Atkinson and Arey, 2003	0.06	
Octane	C ₈ H ₁₈	8.11E-12	GC-FID			Atkinson and Arey, 2003	0.04	
Alkenes and Alkynes								
Ethene	C ₂ H ₄	7.50E-12	GC-FID			IUPAC preferred value	2.26	
Propene	C ₃ H ₆	2.90E-11	GC-FID			IUPAC preferred value	1.11	
trans-2-butene	C ₄ H ₈	6.40E-11	GC-FID			IUPAC preferred value	0.11	
1-Butene	C ₄ H ₈	3.14E-11	GC-FID			IUPAC preferred value	0.26	
1-Pentene	C ₅ H ₁₀	3.14E-11	GC-FID			Atkinson and Arey, 2003	0.10	
Isoprene	C ₅ H ₈	1.00E-10	GC-FID			IUPAC preferred value	0.76	
27.023	C ₂ H ₂	7.50E-13	PTR-ToF-MS	Ethyne		IUPAC preferred value	0.001	
41.0386	C ₃ H ₄	6.95E-13	PTR-ToF-MS	Propyne; 1,2-Propadiene	average	Warnatz, 1984; Atkinson and Arey, 2003	0.27	
67.0543	C ₅ H ₆	5.16E-11	PTR-ToF-MS	1,3-Cyclopentadiene; 3-penten-2-yne	average	Grosjean and Williams, 1992; Boodaghians et al., 1987	0.35	

69.0699	C ₅ H ₈	5.10E-11	PTR-ToF-MS	1-Pentyne; 1,2-Pentadiene; 1,4-Pentadiene; 1,3-Pentadiene; Cyclopentene	average	Boodaghians et al., 1987; Rogers, 1989; Ohta, 1983; Atkinson and Arey, 2003	1.92	C ₅ dienes. Isoprene measured by GC-FID was subtracted
83.0856	C ₆ H ₁₀	8.69E-11	PTR-ToF-MS	Cyclohexene, 1-methyl-cyclopentene; 2,3-dimethylbutadiene; 2-methyl-pentadiene; hexyne; 2-methyl-1,4-pentadiene; 3-methyl-1,3-pentadiene; 4-methyl-1,3-pentadiene; 1,2-hexadiene; 2-4-hexadiene; 1,4-hexadiene; 1,3-hexadiene; 1,5-Hexadiene	average	Atkinson and Arey, 2003; Boodaghians et al., 1987	3.13	C ₆ dienes
85.1012	C ₆ H ₁₂	6.63E-11	PTR-ToF-MS	2-Methyl-2-pentene; 2-Methyl-1-pentene; 3-Methyl-2-pentene; 2-Ethyl-1-butene; 2,3-Dimethyl-2-butene; 1-Hexene; 2-Hexene; 3-Hexene; 4-Methyl-2-pentene	average	Atkinson and Arey, 2003 Ohta, 1983; Grosjean and Williams, 1992	0.57	C ₆ alkenes
95.0856	C ₇ H ₁₀	4.68E-11	PTR-ToF-MS	2-Norbornene		Atkinson et al., 1983	0.76	
97.1012	C ₇ H ₁₂	7.98E-11	PTR-ToF-MS	1-methyl-cyclohexene; cycloheptene; 2-methyl-1,5-hexadiene; 2,4-dimethyl-1,3-pentadiene	average	Aschmann et al., 2012; Atkinson and Arey, 2003; Grosjean and Williams, 1992	2.27	C ₇ dienes
109.1012	C ₈ H ₁₂	1.07E-10	PTR-ToF-MS	Bicyclo[2.2.2]oct-2-ene; 1,2,3-Trimethylcyclopentadiene	average	Atkinson and Arey, 2003; Grosjean and Williams, 1992	3.82	
111.1169	C ₈ H ₁₄	1.20E-10	PTR-ToF-MS	Z-Cyclooctene; 2,5-Dimethyl-2,4-hexadiene; 2,5-Dimethyl-1,5-hexadiene		Aschmann et al., 2012; Atkinson and Arey, 2003	1.89	C ₈ dienes
137.1325	C ₁₀ H ₁₆	3.73E-10	PTR-ToF-MS	1,3,7-Octatriene; α-Pinene	average	IUPAC preferred value; Peeters et al., 2007	3.07	
205.1951	C ₁₅ H ₂₄	1.01E-10	PTR-ToF-MS	β-caryophyllene; α-copaene; α-cedrene; α-humulene; E-β-farnesene; Longifolene		Atkinson and Arey, 2003; IUPAC preferred values	1.10	Sesquiterpenes
Aromatic Compounds								
m- and p-Xylene	C ₈ H ₁₀	1.87E-11	GC-FID		average	Atkinson and Arey, 2003	0.26	

69.0335	C ₄ H ₄ O	4.04E-11	PTR-ToF-MS	Furan		Atkinson, 1986	0.28	
Benzene	C ₆ H ₆	1.20E-12	GC-FID			IUPAC preferred value	0.03	
80.0495	C ₅ H ₅ N	5.31E-13	PTR-ToF-MS	Pyridine		Yeung and Elrod, 2003	0.002	
83.0492	C ₅ H ₆ O	9.00E-11	PTR-ToF-MS	3-methyl-furan		Atkinson et al., 1989	0.79	
Toluene	C ₇ H ₈	5.60E-12	GC-FID			IUPAC preferred value	0.23	
94.0652	C ₆ H ₇ N	5.68E-11	PTR-ToF-MS	Aniline; 4-methyl-pyridine; 3-methyl-pyridine; 2-methyl-pyridine	average of methylpyridines and aniline rate coefficients	Witte et al., 1986; Yeung and Elrod, 2003	0.32	
95.0492	C ₆ H ₆ O	2.80E-11	PTR-ToF-MS	Phenol		IUPAC preferred value	0.87	
97.0648	C ₆ H ₈ O	1.25E-10	PTR-ToF-MS	2,5-dimethylfuran		Aschmann et al., 2011	0.86	
105.0731	C ₈ H ₈	5.80E-11	PTR-ToF-MS	Styrene		Atkinson and Arey, 2003	0.66	
107.0492	C ₇ H ₆ O	1.26E-11	PTR-ToF-MS	Benzaldehyde		IUPAC preferred value	0.23	
107.0855	C ₈ H ₁₀	1.36E-11	PTR-ToF-MS	o-Xylene		Atkinson and Arey, 2003	0.35	m- and p-Xylene mixing ratios measured by GC-FID were subtracted
108.0808	C ₇ H ₉ N	9.60E-12	PTR-ToF-MS	2,6-dimethylpyridine; 2,4-dimethylpyridine; 2,3-dimethylpyridine; 2,5-dimethylpyridine	average	Yeung and Elrod, 2003	0.20	
119.0856	C ₉ H ₁₀	5.95E-11	PTR-ToF-MS	α -methylstyrene; β -methylstyrene	average	Bignozzi et al., 1981	0.72	
121.0648	C ₈ H ₈ O	1.61E-11	PTR-ToF-MS	4-methylbenzaldehyde; 3-methylbenzaldehyde; 2-methylbenzaldehyde; acetophenone	average	Clifford et al., 2005; Aschmann et al., 2010	0.20	
121.1012	C ₉ H ₁₂	5.70E-11	PTR-ToF-MS	1,3,5-trimethylbenzene		Atkinson and Arey, 2003	0.91	
123.0441	C ₇ H ₆ O ₂	1.20E-12	PTR-ToF-MS	benzoic acid	rate coefficient of benzene	IUPAC preferred value	0.005	
125.0961	C ₈ H ₁₂ O	4.04E-11	PTR-ToF-MS	2,3,4,5-tetramethylfuran	rate coefficient of furan	Atkinson, 1986	0.14	
129.0699	C ₁₀ H ₈	2.30E-11	PTR-ToF-MS	Naphthalene		Atkinson and Arey, 2003	0.07	
131.0856	C ₁₀ H ₁₀	2.30E-11	PTR-ToF-MS	Dihydronaphthalene	rate coefficient of naphthalene	Atkinson and Arey, 2003	0.07	

133.1012	C ₁₀ H ₁₂	3.37E-11	PTR-ToF-MS	Tetralin; 2-methyl-1-propenylbenzene	average	Atkinson and Arey, 2003; Chiorboli et al., 1983	0.19	
135.0805	C ₉ H ₁₀ O	3.0E-11	PTR-ToF-MS	Dimethylbenzaldehyde	average	Tse et al., 1997; Aschmann et al., 2010	0.18	
135.1169	C ₁₀ H ₁₄	3.70E-11	PTR-ToF-MS	tert-butylbenzene; p-cymene; 1,2,3,5-tetramethylbenzene; 1,2,3,4-tetramethylbenzene	average	Alarcón et al., 2015; Atkinson and Arey, 2003	1.02	
143.0856	C ₁₁ H ₁₀	5.08E-11	PTR-ToF-MS	2-methyl-Naphthalene; 1-methyl-naphthalene	average	Atkinson and Aschmann, 1986; Phousongphouang and Arey, 2002	0.94	
147.1169	C ₁₁ H ₁₄	5.80E-11	PTR-ToF-MS	4-isopropyl styrene; 3-pentenyl benzene	rate coefficient of styrene	Atkinson and Arey, 2003	1.15	
149.1325	C ₁₁ H ₁₆	1.10E-10	PTR-ToF-MS	Pentamethylbenzene		Alarcón et al., 2015	2.23	
161.1325	C ₁₂ H ₁₆	5.80E-11	PTR-ToF-MS	4-tert-butylstyrene	rate coefficient of styrene	Atkinson and Arey, 2003	1.31	
163.1482	C ₁₂ H ₁₈	1.13E-10	PTR-ToF-MS	Hexamethylbenzene		Atkinson and Arey, 2003	1.30	
175.1482	C ₁₃ H ₁₈	3.85E-11	PTR-ToF-MS	1-2,2-dimethyl-1-methylenepropyl-4-methylbenzene; 2-tert-butyl-indan	average of indan and styrene rate coefficients	Atkinson and Arey, 2003	1.02	
189.1638	C ₁₄ H ₂₀	4.60E-11	PTR-ToF-MS	3,5-dimethyl-t-butylstyrene; 1-t-butyltetralin	average of styrene and tetralin rate coefficients	Atkinson and Arey, 2003	0.591	
197.1325	C ₁₅ H ₁₆	4.15E-12	PTR-ToF-MS	1,1'-1,3-propanediylbis-Benzene; 4-1-methylethyl-1,1'-Biphenyl	average of benzene and biphenyl	Atkinson and Arey, 2003; IUPAC preferred value	0.003	
199.1482	C ₁₅ H ₁₈	2.30E-11	PTR-ToF-MS	substituted naphthalenes	rate coefficient of naphthalene	Atkinson and Arey, 2003	0.05	
OVOCs								
Formaldehyde	HCHO	8.50E-12	Aerolaser Hantzsch			IUPAC preferred value	2.33	
33.0335	CH ₃ OH	9.00E-13	PTR-ToF-MS	Methanol		IUPAC preferred value	0.24	
45.0335	C ₂ H ₄ O	1.50E-11	PTR-ToF-MS	Acetaldehyde		IUPAC preferred value	2.21	
47.0128	HCOOH	4.50E-13	PTR-ToF-MS	Formic acid		IUPAC preferred value	0.07	
57.0335	C ₃ H ₄ O	1.13E-11	PTR-ToF-MS	Acrolein		Atkinson and Arey, 2003	1.51	
59.0491	C ₃ H ₆ O	1.80E-13	PTR-ToF-MS	Acetone		IUPAC preferred value	0.16	
61.0285	C ₂ H ₄ O ₂	6.90E-13	PTR-ToF-MS	Acetic acid		IUPAC preferred value	0.13	
63.0441	C ₂ H ₆ O ₂	1.45E-11	PTR-ToF-MS	1,2-ethanediol		IUPAC preferred value	0.18	

65.0233	CH ₄ O ₃	7.10E-12	PTR-ToF-MS	Hydroxymethyl hydroperoxide		Allen et al., 2018	0.14	
71.0491	C ₄ H ₆ O	2.45E-11	PTR-ToF-MS	Methyl vinyl ketone; methacroleine; isoprene hydroxy hydroperoxide	average	IUPAC preferred values; St Clair et al., 2016	0.42	
73.0284	C ₃ H ₄ O ₂	1.30E-11	PTR-ToF-MS	Methyl glyoxal		IUPAC preferred value	0.21	
73.0648	C ₄ H ₈ O	4.87E-11	PTR-ToF-MS	Butanal; 2-methyl-propanal; tetrahydrofuran; 2-butanone (MEK); ethoxy ethene	average	IUPAC preferred value; Moriarty et al., 2003; Audley et al., 1981; Zhou et al., 2006	1.80	C ₄ carbonyls
75.0441	C ₃ H ₆ O ₂	2.85E-12	PTR-ToF-MS	Hydroxyacetone; propanoic acid	average	IUPAC preferred values	0.10	
77.0598	C ₃ H ₈ O ₂	1.62E-11	PTR-ToF-MS	2-methoxyethanol; 1,2-propanediol		IUPAC preferred value, Porter et al., 1997	0.07	
85.0648	C ₅ H ₈ O	4.43E-11	PTR-ToF-MS	Cyclopentanone; E-2-pentenal; 3-methyl-3-buten-2-one; E-2-methyl-2-butenal; 3-methyl-2-butenal; 3-penten-2-one; 1-penten-3-one; 3,6-dihydro-2H-pyran	average	Atkinson and Arey, 2003; Grosjean and Williams, 1992; Jiménez et al., 2009; Davis et al., 2007	0.52	
87.0441	C ₄ H ₆ O ₂	2.40E-11	PTR-ToF-MS	2,3-Butanedione; acetic acid ethenyl ester; 2-propenoic acid methyl ester; 4-hydroxy-2-butenal	average	Baker et al., 2005a; Teruel et al., 2006; Blanco et al., 2009; Dagaut et al., 1988	0.61	C ₄ dioxygenates
87.0805	C ₅ H ₁₀ O	4.63E-11	PTR-ToF-MS	Pentanal; 3-methyl-2-butanone; 3-pantanone; 2-pentanone; 1-vinyloxypropane; 2-Methyl-3-buten-2-ol MBO; 3-methyl-3-buten-1-ol; 2-methylbutanal; 3-methylbutanal; 1-penten-3-ol; Z-2-penten-1-ol	average	Zhou et al., 2006; IUPAC; Cometto et al., 2008; Atkinson and Arey, 2003; Orlando et al., 2001	2.48	
89.0234	C ₃ H ₄ O ₃	1.20E-13	PTR-ToF-MS	Pyruvic acid		IUPAC preferred value	0.001	
89.0598	C ₄ H ₈ O ₂	6.47E-12	PTR-ToF-MS	1,4-dioxane; formic acid 1-methylethyl ester; ethyl acetate; butyric acid; acetoin; 1-hydroxy-3-butanone	average	Picquet et al., 1998; Aschmann et al., 2000; IUPAC; Moriarty et al., 2003; Pimentel et al., 2010	0.10	
91.039	C ₃ H ₆ O ₃	9.85E-13	PTR-ToF-MS	carbonic acid dimethyl ester; methoxymethyl formate	average	Katrib et al., 2002; O'Donnell et al., 2004	0.003	
99.0805	C ₆ H ₁₀ O	4.76E-11	PTR-ToF-MS	Cyclohexanone; 5-hexen-2-	average	Atkinson and Arey, 2003;	0.59	

				one; z-3-methyl-3-penten-2-one; 4-methyl-3-penten-2-one; E-2-hexenal; Z-3-hexenal; 2-ethyl-2-butenal; E-4-hexene-3-one		Grosjean and Williams, 1992; Wang et al., 2010; Gao et al., 2009; Xing et al., 2012		
101.0598	C ₅ H ₈ O ₂	3.17E-11	PTR-ToF-MS	Methyl methacrylate; E-2-butenoic acid methyl ester; acetylacetone; 2-propenoic acid ethyl ester; glutaraldehyde; vinyl propionate; isopropenyl acetate; n-propenyl acetate	average	Teruel et al., 2006; Teruel et al., 2012; Rogers, 1989; Ferrari et al., 1996; Dagaut et al., 1988	1.52	C ₅ dioxygenates
101.0961	C ₆ H ₁₂ O	4.11E-11	PTR-ToF-MS	Oxepane; 2-hexanone; hexanal; t-butyl vinyl ether; 2-methylpentanal; 3-methylpentanal; 4-methylpentanal; 3,3-dimethylbutanal; 2-ethylbutanal	average	Moriarty et al., 2003; Atkinson and Arey, 2003; Zhou et al., 2012; Le Calvé et al., 1998	0.95	C ₆ oxygenates/ carbonyls
103.0754	C ₅ H ₁₀ O ₂	8.71E-12	PTR-ToF-MS	Butanoic acid methyl ester; n-propyl acetate; 1-hydroxy-2-methyl-3-butanone; 1-hydroxy-3-methyl-2-butanone; 1,2-dioxepane; 5-hydroxy-2-pentanone	average	Schütze et al., 2010; Ferrari et al., 1996; Moriarty et al., 2003; Aschmann et al., 2000; Baker et al., 2005b	0.14	
113.0598	C ₆ H ₈ O ₂	5.80E-11	PTR-ToF-MS	E-hexendione; Z-hexendione	average	Tuazon et al., 1985	0.23	
113.0961	C ₇ H ₁₂ O	2.52E-11	PTR-ToF-MS	E-2-Hepten-1-al; cycloheptanone	average	Davis et al., 2007; Atkinson and Arey, 2003	0.16	
115.0754	C ₆ H ₁₀ O ₂	3.40E-11	PTR-ToF-MS	2,5-hexanedione; ethyl-2-butenoate; methacrylic acid ethyl ester	average	Dagaut et al., 1988; Teruel et al., 2012; Blanco et al., 2006	0.97	
117.0911	C ₆ H ₁₂ O ₂	7.42E-12	PTR-ToF-MS	4-hydroxy-3-hexanone; acetic acid butyl ester; butanoic acid ethyl ester; methyl valerate	average	Schütze et al., 2010; Aschmann et al., 2000; Veillerot et al., 1996; Ferrari et al., 1996	0.13	
127.1118	C ₈ H ₁₄ O	9.88E-11	PTR-ToF-MS	E-2-octenal; cyclooctanone; 6-methyl-5-hepten-2-one	average	Gao et al., 2009; Smith et al., 1996	1.87	

99.0053	<chem>CF3CHO</chem>	1.50E-11	PTR-ToF-MS	Trifluoroacetaldehyde	rate coefficient of acetaldehyde	IUPAC preferred value	0.16	
101.0209	<chem>C2H3F3O</chem>	1.23E-13	PTR-ToF-MS	2,2,2-trifluoroethanol		Sellevåg et al., 2004	0.002	
113.0153	<chem>C6H5Cl</chem>	6.02E-13	PTR-ToF-MS	Chlorobenzene		Bryukov et al., 2009	0.003	
129.0102	<chem>C6H5ClO</chem>	1.32E-11	PTR-ToF-MS	2-chlorophenol; 3-chlorophenol; 4-chlorophenol	average	Kiliç et al., 2007	0.07	
NO_x								
Nitric oxide	NO	9.70E-12	Two-channel CLD			IUPAC preferred value	29.3	
Nitrogen dioxide	NO ₂	9.80E-12	Two-channel CLD			IUPAC preferred value	6.94	
Other inorganic compounds								
Ozone	O ₃	7.30E-14	UV Photometric O ₃ Analyzer			IUPAC preferred value	0.26	
Nitrous acid	HONO	6.00E-12	Long Path Absorption Photometer			IUPAC preferred value	0.13	
Sulfur dioxide	SO ₂	9.30E-13	Chemical Ionization Mass Spectrometer			IUPAC preferred value	0.33	
Carbon monoxide	CO	1.44E-13	Picarro Cavity ring-down spectroscopy instrument			IUPAC preferred value	1.16	

References for reaction rate coefficients

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