Electronic Supplementary Material (ESI) for Environmental Science: Processes & Impacts.

Supplementary Information - Indoor Cooking and Cleaning as a Source of Outdoor Air Pollution in Urban Environments

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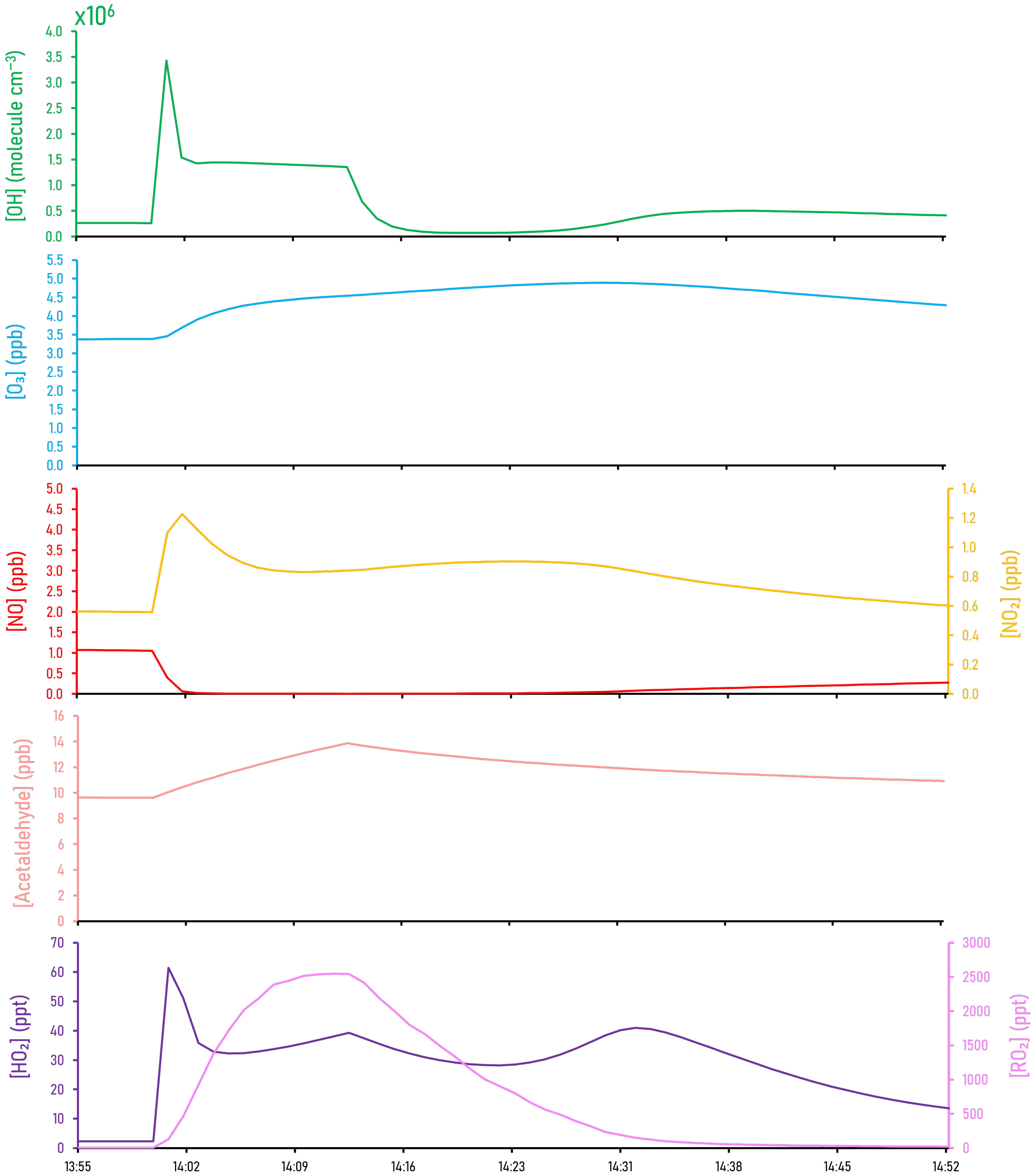

Figure S1: The concentrations of key indoor species during a 13-minute cleaning event (starting at 2pm).

Table S1: The primary emission rates of species from indoor wooden and painted surfaces in a kitchen, calculated from Plaisance *et al.* (2017); Alapieti *et al.* (2021); Cheng *et al.* (2015). These emission rates are constant and have been added to INCHEM-Py.

Species	**Emission Rate (molecule cm^{-3} s^{-1})**
Formaldehyde	1.7 x 10^8
Acetaldehyde	9.6 x 10^7
Propanal	4.1 x 10^7
Butanal	5.0 x 10^7
Pentanal	4.1 x 10^7
Hexanal	1.1 x 10^8
Heptanal	5.2 x 10^6
Octanal	4.8 x 10^6
Nonanal	8.5 x 10^6
Decanal	4.5 x 10^6

Table S2: The near-field concentrations ($C_{\mathrm{i,nf}}$) included in INCHEM-Py.

Species		
Formaldehyde	Acetaldehyde	Propanal
3-Methylbutanal	Acrolein	Methacrolein
Crotonaldehyde	Pentanal	Hexanal
Heptanal	Octanal	Nonanal
Decanal	2-Nonenal	Acetone
2-Butanone (MEK)	3-Buten-2-one (MVK)	Cyclohexanone
Benzaldehyde	o-Tolualdehyde	m-Tolualdehyde
p-Tolualdehyde	2,5-Dimethylbenzaldehyde	Benzene
Toluene	p-Xylene	m-Xylene
o-Xylene	Ethylbenzene	Propylbenzene
2-Ethyltoluene	3-Ethyltoluene	4-Ethyltoluene
1,3,5-Trimethylbenzene	1,2,4-Trimethylbenzene	1,2,3-Trimethylbenzene
p-Dichlorobenzene	Styrene	Cumene
Phenol	Ethane	Propane
Butane	Isobutane	2,2-Dimethylbutane
2,3-Dimethylbutane	Pentane	2-Methylpentane
3-Methylpentane	Isopentane	Hexane
2-Methylhexane	3-Methylhexane	Heptane
Octane	Nonane	Decane
Undecane	Dodecane	Cyclohexane
Ethene	Propene	1-Butene
cis-2-Butene	trans-2-Butene	2-Methyl-1-butene
2-Methyl-2-butene	Isoprene	1,3-Butadiene

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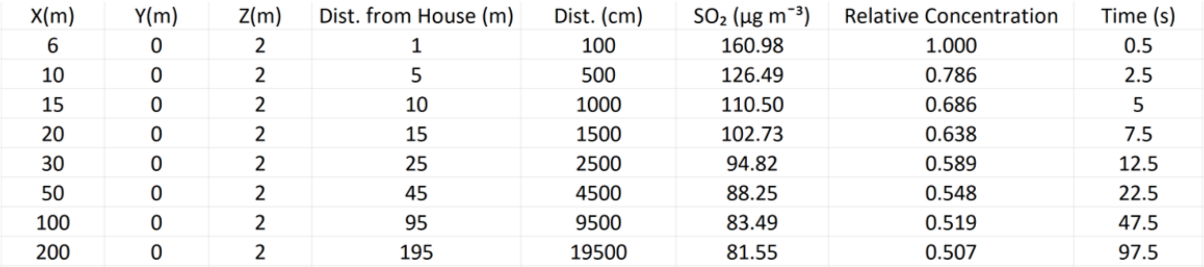
Species		
trans-2-Pentene	cis-2-Pentene	Ethyne
Methanol	Ethanol	Isopropanol
1-Propanol	1-Butanol	1-Pentanol
1-Hexanol	2-Butoxyethanol	Linalool
Chloroform	Methylchloroform	Dichloromethane
Trichloroethylene	Tetrachloroethylene	1,2-Dichloroethane
Hydrogen Chloride	Chloromethane	α-Pinene
β-Pinene	Limonene	Δ3-Carene
Camphene	Formic Acid	Acetic Acid
Propanoic Acid	Butanoic Acid	Pentanoic Acid
Heptanoic Acid	Hydrogen Peroxide	β-Caryophyllene
Methane (CH_4)	Carbon Monoxide (CO)	Sulfur Dioxide (SO_2)
Nitric Acid (HNO_3)	Peroxyacetyl Nitrates (PAN)	-

Table S3: The resultant grid from the ADMS output. The relative concentration and time columns have been added to the grid. The chosen distance was 95 m from which the loss rate was calculated in Equation S1 below.



X(m)	Y(m)	Z(m)	Dist. from House (m)	Dist. (cm)	SO_2 ($\mu g\ m^{-3}$)	Relative Concentration	Time (s)
6	0	2	1	100	160.98	1.000	0.5
10	0	2	5	500	126.49	0.786	2.5
15	0	2	10	1000	110.50	0.686	5
20	0	2	15	1500	102.73	0.638	7.5
30	0	2	25	2500	94.82	0.589	12.5
50	0	2	45	4500	88.25	0.548	22.5
100	0	2	95	9500	83.49	0.519	47.5
200	0	2	195	19500	81.55	0.507	97.5

Equation S1: Calculations for determining loss rate (s^{-1}) using the ADMS grid (Table S3). A loss rate of 1.09 x 10^{-2} s^{-1} was calculated and used in the near-field framework. This value was used as a dilution factor for the near-field concentration because we explored street lengths varying between 100 and 140 m. The loss rates calculated over the whole street was used in our ten-house analysis.

$$\frac{1}{time\ (s)} \times relative\ concentration = loss\ rate\ (s^{-1})$$

$$\frac{1}{47.5\ s} \times 0.519 = 1.09 \times 10^{-2}\ s^{-1}$$

Table S4: The emission rates (calculated from HOMEChem (Farmer *et al.*, 2019)), in molecule cm^{-3} s^{-1}, inputted into INCHEM-Py for the breakfast, lunch, dinner and cleaning activities.

Species	**Breakfast**	**Lunch**	**Dinner**	**Cleaning**
Methane	-	6.9×10^{8}	2.0×10^{8}	-
Carbon Monoxide	$1.9\ \times10^{10}$	2.8×10^{10}	1.6×10^{10}	-
Acetaldehyde	$9.8\ \times10^{6}$	1.9×10^{7}	3.6×10^{7}	1.8×10^{8}
Acetone	-	-	-	1.8×10^{7}
α-Pinene	2.9×10^{6}	-	-	2.2×10^{5}
Benzene	7.2×10^{5}	5.8×10^{5}	3.8×10^{5}	1.8×10^{6}
β-Pinene	2.6×10^{6}	-	2.0×10^{6}	1.3×10^{7}
Chloroform	-	-	-	3.0×10^{6}
cis-But-2-ene	6.6×10^{4}	7.2×10^{4}	1.8×10^{4}	9.5×10^{4}
cis-Pent-2-ene	2.2×10^{4}	2.5×10^{4}	3.1×10^{4}	-
Ethane	2.9×10^{7}	4.6×10^{7}	2.4×10^{8}	-
Ethene	6.2×10^{7}	4.1×10^{7}	2.9×10^{7}	1.6×10^{8}
Ethylbenzene	1.2×10^{6}	-	-	1.3×10^{5}
Ethyne	3.3×10^{7}	3.9×10^{7}	7.8×10^{7}	3.8×10^{8}
Isoprene	1.5×10^{6}	1.6×10^{6}	3.8×10^{6}	1.8×10^{7}
Isobutane	2.0×10^{7}	2.4×10^{7}	4.1×10^{8}	-
Isopentane	9.8×10^{5}	-	-	-
Limonene	8.3×10^{6}	-	2.9×10^{6}	1.5×10^{7}
Methyl Ethyl Ketone (MEK)	1.2×10^{6}	8.2×10^{5}	-	-
m-Xylene	5.2×10^{4}	-	8.8×10^{4}	6.7×10^{5}
p-Xylene	5.2×10^{4}	-	8.8×10^{4}	6.7×10^{5}
Butane	1.4×10^{7}	9.0×10^{6}	-	-

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Species	**Breakfast**	**Lunch**	**Dinner**	**Cleaning**
Decane	1.1×10^5	-	9.9×10^4	4.5×10^5
Heptane	1.8×10^5	-	1.4×10^5	6.8×10^5
Hexane	1.4×10^6	1.9×10^6	-	-
Nonane	1.7×10^6	3.7×10^4	1.4×10^5	-
Octane	1.4×10^5	1.8×10^5	1.8×10^4	1.3×10^5
Pentane	1.1×10^6	1.1×10^6	2.1×10^6	1.0×10^7
Propane	3.4×10^9	3.7×10^9	-	-
Propene	5.9×10^7	3.9×10^7	2.9×10^7	1.4×10^8
Styrene	1.6×10^5	2.8×10^5	-	5.3×10^5
o-Xylene	1.6×10^5	2.8×10^5	-	5.3×10^5
trans-But-2-ene	5.3×10^4	6.6×10^4	1.9×10^4	1.1×10^5
But-1-ene	4.8×10^5	7.8×10^5	1.3×10^6	-
o-Ethyltoluene	2.8×10^6	-	1.5×10^5	7.5×10^5
2-Methylbut-1-ene	3.4×10^4	-	4.9×10^3	2.7×10^4
m-Ethyltoluene	5.6×10^6	-	-	-
p-Ethyltoluene	5.6×10^6	-	-	-
1,2,3-Trimethylbenzaldehyde	1.9×10^6	-	2.8×10^6	1.5×10^7
1,3,5-Trimethylbenzaldehyde	1.4×10^5	2.4×10^6	9.9×10^4	4.7×10^5
Chloroformic Acid	-	-	-	3.0×10^6
Nitryl Chloride	-	-	-	1.8×10^8
Chlorine (Cl_2)	-	-	-	6.7×10^8
Hypochlorous Acid	-	-	-	5.7×10^8

Table S5: The outdoor concentrations, given in ppb, for the gas-phase species included in the INCHEM-Py model (Uchiyama *et al.*, 2015; Baudic *et al.*, 2016; Lü *et al.*, 2006; Mentese and Bas, 2020; Bari and Kindzierski, 2018; Sturaro *et al.*, 2010; Bari *et al.*, 2016; Gallego *et al.*, 2016; Brickus *et al.*, 1998; Hellén *et al.*, 2018; Hakola *et al.*, 2009; He *et al.*, 2010; Dlugokencky, 2022; Vichi *et al.*, 2016; Liu *et al.*, 2018; Li *et al.*, 2018; EEA, 2018).

Species	**Outdoor Concentration (ppb)**
Formaldehyde	2.5
Acetaldehyde	1.6
Propanal	0.38
3-Methylbutanal	0.04
Acrolein	0.11
Methacrolein	0.11
Crotonaldehyde	0.07
Pentanal	0.10
Hexanal	0.11
Heptanal	0.08
Octanal	0.10
Nonanal	0.60
Decanal	0.16
2-Nonenal	0.05
Acetone	2.0
2-Butanone (MEK)	0.22
3-Buten-2-one (MVK)	0.11
Cyclohexanone	0.69
Benzaldehyde	0.06
o-Tolualdehyde	0.05
m-Tolualdehyde	0.08

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Species	**Outdoor Concentration (ppb)**
p-Tolualdehyde	0.08
2,5-Dimethylbenzaldehyde	0.32
Benzene	0.39
Toluene	1.7
p-Xylene	0.25
m-Xylene	0.25
o-Xylene	0.17
Ethylbenzene	0.36
Propylbenzene	0.16
2-Ethyltoluene	0.01
3-Ethyltoluene	0.02
4-Ethyltoluene	0.01
1,3,5-Trimethylbenzene	0.07
1,2,4-Trimethylbenzene	0.22
1,2,3-Trimethylbenzene	0.05
Styrene	0.09
Cumene	0.12
Phenol	0.71
Ethane	3.7
Propane	1.5
Butane	1.4
Isobutane	0.83
2,2-Dimethylbutane	0.08
2,3-Dimethylbutane	0.11

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Species	**Outdoor Concentration (ppb)**
Pentane	0.35
2-Methylpentane	0.16
3-Methylpentane	0.10
Isopentane	0.60
Hexane	0.45
2-Methylhexane	0.10
3-Methylhexane	0.13
Heptane	0.02
Octane	0.02
Nonane	0.12
Decane	0.40
Undecane	0.59
Dodecane	0.04
Cyclohexane	0.03
Ethene	1.4
Propene	0.37
1-Butene	0.16
cis-2-Butene	0.02
trans-2-Butene	0.02
2-Methyl-1-butene	0.02
2-Methyl-2-butene	0.02
Isoprene	0.09
1,3-Butadiene	0.02
trans-2-Pentene	0.02

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Species	**Outdoor Concentration (ppb)**
cis-2-Pentene	0.01
Ethyne	0.64
Methanol	4.5
Ethanol	6.6
Isopropanol	3.8
1-Propanol	0.51
1-Butanol	1.0
1-Pentanol	0.002
1-Hexanol	0.001
2-Butoxyethanol	1.0
Linalool	0.001
Chloroform	0.03
Methylchloroform	0.31
Dichloromethane	0.10
Trichloroethylene	0.37
Tetrachloroethylene	0.02
1,2-Dichloroethane	0.02
Hydrogen Chloride	1.5
Chloromethane	0.57
α-Pinene	0.13
β-Pinene	0.05
Limonene	0.10
Δ3-Carene	0.11
Camphene	0.02

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Species	**Outdoor Concentration (ppb)**
Formic Acid	7.5
Acetic Acid	15.7
Propanoic Acid	0.08
Butanoic Acid	0.06
Pentanoic Acid	0.03
Heptanoic Acid	0.004
Hydrogen Peroxide	1.3
β-Caryophyllene	0.004
Methane (CH_4)	1891
Carbon Monoxide (CO)	195
Sulfur Dioxide (SO_2)	0.70
Nitric Acid (HNO_3)	0.39
Peroxyacetyl Nitrates (PAN)	2.2

Table S6: The percentage change of key indoor species concentrations following the omission of propane emissions during the layered day simulation. A positive percentage change indicates an increase in concentration, whereas a negative percentage change indicates a decrease in concentration following the removal of propane emissions.

Species	**Percentage Change (%)**
OH	−4.9
O_3	1.9
NO	−0.2
NO_2	2.1
HO_2	−21.3
RO_2	51.8

Equation S2: Calculation for VOC Emissions (Section 3.4 - How Indoor Sources contribute to Outdoor Air Pollution)

$$\frac{C_i \times ACR \times 10^6 \times M_r \times V_{house}}{N_A} = E_i$$

where:

C_i = Indoor VOC Concentration (molecule cm^{-3})

ACR = Air Change Rate (12 day^{-1})

M_r = Molecular Weight (g mol^{-1})

V_{house} = Volume of a House (500 m^3)

N_A = Avogadro's Constant (6.022×10^{23} molecule mol^{-1})

E_i = VOC Emission Rate (g day^{-1})

The emission rates from the NMVOCs are then summed to produce a total VOC emission rate ($E_{i_{tot}}$).

References

H. Plaisance, J. Vignau-Laulhere, P. Mocho, N. Sauvat, K. Raulin and V. Desauziers, Volatile organic compounds concentrations during the construction process in newly-built timber-frame houses: Source identification and emission kinetics, *Environmental Science: Processes and Impacts*, 2017, **19**, 696–710.

T. Alapieti, E. Castagnoli, L. Salo, R. Mikkola, P. Pasanen and H. Salonen, The effects of paints and moisture content on the indoor air emissions from pinewood (Pinus sylvestris) boards, *Indoor Air*, 2021, **31**, 1563–1576.

Y. H. Cheng, C. C. Lin and S. C. Hsu, Comparison of conventional and green building materials in respect of VOC emissions and ozone impact on secondary carbonyl emissions, *Building and Environment*, 2015, **87**, 274–282.

D. K. Farmer, M. E. Vance, J. P. Abbatt, A. Abeleira, M. R. Alves, C. Arata, E. Boedicker, S. Bourne, F. Cardoso-Saldaña, R. Corsi, P. F. Decarlo, A. H. Goldstein, V. H. Grassian, L. Hildebrandt Ruiz, J. L. Jimenez, T. F. Kahan, E. F. Katz, J. M. Mattila, W. W. Nazaroff, A. Novoselac, R. E. O'Brien, V. W. Or, S. Patel, S. Sankhyan, P. S. Stevens, Y. Tian, M. Wade, C. Wang, S. Zhou and Y. Zhou, Overview of HOMEChem: House Observations of Microbial and Environmental Chemistry, *Environmental Science: Processes and Impacts*, 2019, **21**, 1280–1300.

S. Uchiyama, T. Tomizawa, A. Tokoro, M. Aoki, M. Hishiki, T. Yamada, R. Tanaka, H. Sakamoto, T. Yoshida, K. Bekki, Y. Inaba, H. Nakagome and N. Kunugita, Gaseous chemical compounds in indoor and outdoor air of 602 houses throughout Japan in winter and summer, *Environmental Research*, 2015, **137**, 364–372.

A. Baudic, V. Gros, S. Sauvage, N. Locoge, O. Sanchez, R. Sarda-Estève, C. Kalogridis, J. E. Petit, N. Bonnaire, D. Baisnée, O. Favez, A. Albinet, J. Sciare and B. Bonsang, Seasonal variability and source apportionment of volatile organic compounds (VOCs) in the Paris megacity (France), *Atmospheric Chemistry and Physics*, 2016, **16**, 11961–11989.

H. Lü, S. Wen, Y. Feng, X. Wang, X. Bi, G. Sheng and J. Fu, Indoor and outdoor carbonyl compounds and BTEX in the hospitals of Guangzhou, China, *Science of the Total Environment*, 2006, **368**, 574–584.

S. Mentese and B. Bas, A year-round motoring of ambient volatile organic compounds across Dardanelles strait, *Journal of Chemical Metrology*, 2020, **14**, 177–189.

M. A. Bari and W. B. Kindzierski, Ambient volatile organic compounds (VOCs) in Calgary, Alberta: Sources and screening health risk assessment, *Science of the Total Environment*, 2018, **631-632**, 627–640.

A. Sturaro, R. Rella, G. Parvoli and D. Ferrara, Long-term phenol, cresols and BTEX monitoring in urban air, *Environmental Monitoring and Assessment*, 2010, **164**, 93–100.

M. A. Bari, W. B. Kindzierski and D. Spink, Twelve-year trends in ambient concentrations of volatile organic compounds in a community of the Alberta Oil Sands Region, Canada, *Environment International*, 2016, **91**, 40–50.

E. Gallego, F. J. Roca, J. F. Perales, X. Guardino, E. Gadea and P. Garrote, Impact of formaldehyde and VOCs from waste treatment plants upon the ambient air nearby an urban area (Spain), *Science of the Total Environment*, 2016, **568**, 369–380.

L. S. Brickus, J. N. Cardoso and F. R. De Aquino Neto, Distributions of indoor and outdoor air pollutants in Rio de Janeiro, Brazil: Implications to indoor air quality in bayside offices, *Environmental Science and Technology*, 1998, **32**, 3485–3490.

H. Hellén, A. P. Praplan, T. Tykkä, I. Ylivinkka, V. Vakkari, J. Bäck, T. Petäjä, M. Kulmala and H. Hakola, Long-term measurements of volatile organic compounds highlight the importance of sesquiterpenes for the atmospheric chemistry of a boreal forest, *Atmospheric Chemistry and Physics*, 2018, **18**, 13839–13863.

H. Hakola, H. Hellén, V. Tarvainen, J. Bäck, J. Patokoski and J. Rinne, Annual variations of atmospheric VOC concentrations in a boreal forest, *Boreal Environment Research*, 2009, **14**, 722–730.

S. Z. He, Z. M. Chen, X. Zhang, Y. Zhao, D. M. Huang, J. N. Zhao, T. Zhu, M. Hu and L. M. Zeng, Measurement of atmospheric hydrogen peroxide and organic peroxides in Beijing before and during the 2008 Olympic Games: Chemical and physical factors influencing their concentrations, *Journal of Geophysical Research*, 2010, **115**, D17307.

E. Dlugokencky, *NOAA/GML CH4 Trends (Date Accessed: March 2022)*, 2022, `https://gml.noaa.gov/webdata/ccgg/trends/ch4/ch4_mm_gl.txt`.

F. Vichi, L. Mašková, M. Frattoni, A. Imperiali and J. Smolík, Simultaneous measurement of nitrous acid, nitric acid, and nitrogen dioxide by means of a novel multipollutant diffusive sampler in libraries and archives, *Heritage Science*, 2016, **4**, 1–8.

L. Liu, X. Wang, J. Chen, L. Xue, W. Wang, L. Wen, D. Li and T. Chen, Understanding unusually high levels of peroxyacetyl nitrate (PAN) in winter in Urban Jinan, China, *Journal of Environmental Sciences (China)*, 2018, **71**, 249–260.

M. Li, E. Karu, C. Brenninkmeijer, H. Fischer, J. Lelieveld and J. Williams, Tropospheric OH and stratospheric OH and Cl concentrations determined from CH4, CH3Cl, and SF6 measurements, *npj Climate and Atmospheric Science*, 2018, **1**, 1–7.

EEA, *European Air Quality Portal, https://eeadmz1-cws-wp-air02.azurewebsites.net/, (Date Accessed: December 2021)*, 2018.