

## 5. Appendices

### 5.1. Symbols

Symbol	Description	Unit(s)/Value
$AB$	Airbeam signal strength	Hz
$a_x$	Arbitrary multiplication factors	
$A_x$	Intermediate functions	Various
$b$	Velocity calibration exponent	
$B_1, B_2$	Organic signal distribution weighting factors	Hz
$C$	Ambient volumetric mass concentration	$\mu\text{g m}^{-3}$
$CE$	Collection efficiency	
$D^*$	Velocity calibration size constant	nm
$D_a$	Particle classical (transition regime) aerodynamic diameter	nm (or $\mu\text{m}$ )
$D_m$	Particle electromobility diameter	nm (or $\mu\text{m}$ )
$D_o$	Particle optical diameter	nm (or $\mu\text{m}$ )
$D_s$	Particle Stokes diameter	nm (or $\mu\text{m}$ )
$D_v$	Particle volume equivalent diameter	nm (or $\mu\text{m}$ )
$D_{va}$	Particle vacuum (free molecular regime) aerodynamic diameter	nm (or $\mu\text{m}$ )
$e$	Elementary charge	$1.602176 \times 10^{-19} \text{ C}$
$f^x$	Normal distribution function	
$G$	Relative multiplier gain and detection efficiency	
$I$	Detected ion rate	Hz
$IE$	Ionisation efficiency	
$k$	Reaction rate	$\text{s}^{-1}$
$L_c$	Chamber length	m
$m$	Relative molecular mass	
$M$	Size resolved ambient volumetric mass concentration	$\mu\text{g m}^{-3}$
$m/z$	Ion mass to charge ratio	
$\mathbf{M}_s$	Species-specific partial mass spectrum extraction matrix	
$MW$	Molecular weight	$\text{g mol}^{-1}$
$N_A$	Avagadro's number	$6.022142 \times 10^{23} \text{ mol}^{-1}$
$N_I$	Number of ions detected per TOF scan	
$n_v$	Differential volume concentration function	
$p_x$	Relative signal weighting factor	
$Q$	Volumetric flow rate	$\text{cm}^3 \text{ s}^{-1}$
$Q^*$	Flowrate correction offset	$\text{cm}^3 \text{ s}^{-1}$

Symbol	Description	Unit(s)/Value
$q_I$	Electronic noise parameter	Hz s <sup>0.5</sup>
$r$	Pearson's linear covariance	
$RH$	Relative humidity	%
$RIE$	Relative ionisation efficiency	
$S$	Particle Jayne shape factor	
$t$	Temperature	°C
$T$	Relative quadrupole transmission	
$t_d$	Dew point temperature	°C
$t_p$	Particle time of flight	s
$t_q$	Quadrupole time	s
$t_{RH}$	Inlet setpoint temperature	°C
$t_s$	Sampling time	s
$U_1$	Quadrupole voltage DC component	V
$U_2$	Quadrupole voltage AC component	V
$V$	Volume concentration	
$v_g$	Gas velocity after nozzle	m s <sup>-1</sup>
$v_l$	Gas velocity before nozzle	m s <sup>-1</sup>
$v_p$	Particle velocity	m s <sup>-1</sup>
$V_q$	Quadrupole voltage	V
$z$	Relative electronic charge	
$\alpha$	Multiplier error enhancement factor	
$\pi$	Pi	3.141593
$\rho_0$	Unit density	1 g cm <sup>-3</sup>
$\rho_{eff}$	Particle effective density	g cm <sup>-3</sup>
$\rho_p$	Particle physical density	g cm <sup>-3</sup>
$\sigma$	Standard error	Various
$\nu_q$	Quadrupole AC frequency	Hz
$\chi_{v,inv}$	Particle inverse dynamic shape factor	

Table 5.1.I. Mathematical symbols used in this thesis with their units and where appropriate, values.

## 5.2. Acronyms and abbreviations

Acronym	Meaning
AC	Alternating Current
ACE	Aerosol Characterisation Experiment
AL	(NOAA) Aeronomy Laboratory
AMS	(Aerodyne) Aerosol Mass Spectrometer
APCI-MS	Atmospheric Pressure Chemical Ionisation Mass Spectrometry
APS	Aerodynamic Particle Sizer
ARI	Aerodyne Research Incorporated
ASCII	American Standard Code for Information Interchange
ATOFMS	Aerosol Time Of Flight Mass Spectrometer
BADC	British Atmospheric Data Centre
BAe	British Aerospace Engineering
BAM	Beta Attenuation Monitor
BC	Black Carbon
CAART	Chemical Analysis of Aerosols in Real Time
Caltech	California Institute of Technology
CCD	Charge Coupled Device
CCN	Cloud Condensation Nucleus/Nuclei
CEH	Centre for Ecology and Hydrology
CIRPAS	Center for Interdisciplinary Remotely-Piloted Aircraft Studies
CLACE	Cloud and Aerosol Characterisation Experiments
CNR	Consiglio Nazionale delle Ricerche (Italy)
CPC	Condensation Particle Counter
DC	Direct Current
DLR	Deutschen Zentrum für Luft- und Raumfahrt
DMA	Differential Mobility Analyser
DMPS	Differential Mobility Particle Sizer
DMS	Dimethyl Sulphide ((CH <sub>3</sub> ) <sub>2</sub> S)
DOE	Department of Energy (USA)
EC	Elemental Carbon
EI	Electron Impact (ionisation)
FAAM	Facility for Airborne Atmospheric Research
FID	Flame Ionisation Detector
FWHM	Full Width Half Maximum
GC	Gas Chromatography
GMT	Greenwich Mean Time
HDF	Hierarchical Data Format
HTDMA	Hygroscopicity Tandem Differential Mobility Analyser
HYSPLIT	Hybrid Single-Particle Lagrangian Integrated Trajectory

Acronym	Meaning
IC	Ion Chromatography
ID	Internal Diameter
IGAC	International Global Atmospheric Chemistry
ITCT 2K2	Intercontinental Transport and Chemical Transformation 2002
itx	Igor Text
IUPAC	International Union of Pure and Applied Chemistry
JMS	Jump Mass Spectrum
LDI	Laser Desorption and Ionisation
LFV	Lower Fraser Valley
Lovol	Low-volume sampler
MACR	Methacryl Aldehyde ( $\text{CH}_2=\text{C}(\text{CH}_3)\text{CHO}$ )
MBL	Marine Boundary Layer
MBO	2-methyl-3-buten-2-ol ( $\text{CH}_2=\text{CHC}(\text{CH}_3)_2\text{OH}$ )
MEK	Methyl Ethyl Ketone ( $\text{C}_2\text{H}_5\text{COCH}_3$ )
MOUDI	Micro Orifice Uniform Deposit Impactor
MS	Mass Spectrometry
MSA	Methylsulphonic Acid ( $\text{CH}_3\text{SO}_2\text{OH}$ )
MTBE	Methyl Tertiary Butyl Ether ( $\text{tert-C}_4\text{H}_9\text{OCH}_3$ )
MVK	Methyl Vinyl Ketone ( $\text{CH}_2=\text{CHCOCH}_3$ )
NAEI	National Atmospheric Emissions Inventory (UK)
NAMBLEX	North Atlantic Marine Boundary Layer Experiment
NEAQS	New England Air Quality Study
NERC	Natural Environmental Research Council (UK)
NIST	National Institute of Standards and Technology (USA)
NOAA	National Oceanographic and Atmospheric Administration (USA)
nss	Non-sea salt
OC	Organic Carbon
OPC	Optical Particle Counter
PAH	Polycyclic Aromatic Hydrocarbon
PALMS	Particle Analysis by Laser Mass Spectrometry
PAMS	Particle Analysis by quadrupole Mass Spectrometry
PERD	Panel on Energy Research and Development (Canada)
PILS	Particle Into Liquid Sampler
PM	Particulate Matter
PSL	Polystyrene Latex
PST	Pacific Summer Time
PTFE	Polytetrafluoroethylene ( $\text{C}_2\text{F}_2$ ) <sub>n</sub>
PVC	Polyvinyl Chloride ( $\text{C}_2\text{H}_3\text{Cl}$ ) <sub>n</sub>
PVF	Polyvinyl Fluoride ( $\text{C}_2\text{H}_3\text{F}$ ) <sub>n</sub>

<b>Acronym</b>	<b>Meaning</b>
QUEST	Quantification of Aerosol Nucleation in the European Boundary Layer
RF	Radio Frequency
RH	Relative Humidity
RSMS	Rapid Single-particle Mass Spectrometry
SASUA	Sources And Sinks of Urban Aerosol
SMPS	Scanning Mobility Particle Sizer
SP2	Single Particle Soot Photometer
TDPBMS	Thermal Desorption Particle Beam Mass Spectrometer
TEC	Thermoelectric Cooler
TEOM	Tapered Element Oscillating Microbalance
TOF	(particle) Time Of Flight
ToF-AMS	Time of Flight Aerosol Mass Spectrometer
TORCH	Tropospheric Organic Chemistry
uCPC	Ultrafine Condensation Particle Counter
UMIST	University of Manchester Institute of Science and Technology
URGENT	Urban Regeneration and the Environment
UTC	Universal Time (GMT)
UV	Ultra Violet
VOC	Volatile Organic Carbon
WSOC	Water-Soluble Organic Carbon
YAG	Yttrium Aluminium Garnet

Table 5.2.I. Acronyms and abbreviations used in this thesis with their meanings.

### 5.3. Calculation parameters

Experiment	Relative Ionisation Efficiencies				TOF Organic Weighting Factors				
	$RIE_{NO_3}$	$RIE_{SO_4}$	$RIE_{NH_4}$	$RIE_{Org}$	$P_{43}$	$P_{44}$	$P_{55}$	$P_{57}$	$P_{69}$
SASUA 3	1.1	1.15	3.5	1.4	2.36		1.6		2.35
Man. Jan.	1.1	1.15	5	1.4	5.84		4	2.89	
Man. Jun.	1.1	1.15	3.5	1.4	4		3.7	1	
Pacific 2001	1.1	1.15	3.5	1.4	3.2		3.09	2.27	
ACE-Asia	1.1	1.15	3.5	1.4	5.9		5.67	-0.83	
ITCT 2K2	1.1	1.15	4.9	1.4	2.93	2.75	2.75		2.49
NAMBLEX	1.1	1.15	3.5	1.4	7.8		3.4	2.6	

Table 5.3.I. The parameters used for the processed data presented in this thesis, identified by the project name. The application of relative ionisation efficiencies is described in section 2.4.2.4., while the organic weighting factors is described in section 2.4.3.2.

## 5.4. Fragmentation tables

<i>m/z</i>	frag_air	frag_nitrate	frag_K	frag_chloride
14	[14], -frag_nitrate[14]	0.04*frag_nitrate[30] <sup>b</sup> , 0.04*frag_nitrate[46] <sup>b</sup>		
15	0.00368*frag_air[14] <sup>a</sup>			
16	Frag_O16[16], frag_RH[16]			
17	0.000391*frag_O16[16] <sup>a</sup> , frag_RH[17]			
18	0.002*frag_O16[16] <sup>a</sup> , frag_RH[18]			
19	frag_RH[19]			
20	[20], -frag_organic[20], -frag_sulphate[20], -frag_water[20]			
...				
28	[28]			
29	0.00736*frag_air[28] <sup>a</sup>			
30	0.0000136*frag_air[28] <sup>a</sup>	[30], -frag_air[30], -frag_organic[30]		
31		0.00405*frag_nitrate[30] <sup>a</sup>		
32	[32], -frag_sulphate[32], -frag_nitrate[32]	0.002*frag_nitrate[30] <sup>a</sup>		
33	0.000763*frag_air[32] <sup>a</sup>			
34	0.00402*frag_air[32] <sup>a</sup>			
35				[35]
36	0.00338*frag_air[40] <sup>a</sup>			[36], -frag_air[36]
37				0.323*frag_chloride[35] <sup>a</sup>
38	0.000633*frag_air[40] <sup>a</sup>			0.323*frag_chloride[36] <sup>a</sup>
39			[39]	
40	[40]			
41			0.0722*frag_K[39] <sup>a</sup>	
...				
44	0.000734*frag_air[28] <sup>b</sup>			
45				
46		[46]		
47		0.00443*frag_nitrate[46] <sup>a</sup>		
48		0.004*frag_nitrate[46] <sup>a</sup>		
...				
63		0.003*frag_nitrate[30] <sup>b</sup> , 0.002*frag_nitrate[46] <sup>b</sup>		

Table 5.4.I. The fragmentation tables for the chemical components of air (frag\_air), particulate nitrate (frag\_NO3), potassium (frag\_K) and chloride (frag\_Cl). Contributions marked with an <sup>a</sup> use multipliers based on predicted contributions from common isotopes. Those marked with a <sup>b</sup> are based on the analysis of laboratory and field studies, which will be presented in future publications. The entry at frag\_air[44] is an estimate of the gas-phase CO<sub>2</sub> contribution to the mass spectrum. This may require tweaking depending on the ambient concentration during an experiment. Omitted rows

are blank. Note that all contributions from the  $^{37}\text{Cl}$  isotope are calculated based on signals due to  $^{35}\text{Cl}$ .

<i>m/z</i>	frag_RH	frag_O16	frag_water	frag_NH4
15				0.1*frag_NH4[16] <sup>b</sup>
16	0.04*frag_RH[18] <sup>b</sup>	0.353*frag_air[14] <sup>b</sup>	0.04*frag_water[18] <sup>b</sup>	[16], -frag_water[16], -frag_air[16], -frag_sulphate[16], -frag_organic[16]
17	0.25*frag_RH[18] <sup>b</sup>		0.25*frag_water[18] <sup>b</sup>	[17], -frag_water[17], -frag_air[17], -frag_sulphate[17], -frag_organic[17]
18	0.01*frag_air[28] <sup>b</sup>		[18], -frag_air[18], -frag_sulphate[18], -frag_organic[18]	
19	0.000691*frag_RH[18] <sup>a</sup> , 0.002*frag_RH[17] <sup>a</sup>		0.000691*frag_water[18] <sup>a</sup> , 0.002*frag_water[17] <sup>a</sup>	
20	0.002*frag_RH[18] <sup>a</sup>		0.002*frag_water[18] <sup>a</sup>	

Table 5.4.II. The fragmentation tables for gas phase water (frag\_RH), the  $^{16}\text{O}^+$  and  $^{16}\text{O}_2^{++}$  fragments due to gas phase oxygen (frag\_O16), particulate water (frag\_water) and ammonium (frag\_NH4). The relative contribution to the gas phase from water vapour is typically variable in time and to be completely thorough, should be accounted for by adjusting the multiplier in the frag\_RH[18] entry. However, any inaccuracy in this number will only affect the ability to quantitatively distinguish water vapour from particulate water, which is not required in most applications. The ratio governing the  $^{16}\text{O}^+$  and  $^{16}\text{O}_2^{++}$  peak is variable between instruments and configurations and has to be set correctly in order to be able to calculate ammonium concentrations correctly.

<i>m/z</i>	frag_sulphate	frag_H2SO4	frag_SO3	frag_organic
12				[12]
13				[13]
14				
15				[15], -frag_NH4[15], -frag_air[15]
16	frag_SO3[16]		0.04*frag_SO3[18] <sup>b</sup>	0.04*frag_organic[18] <sup>b</sup>
17	frag_SO3[17]		0.25*frag_SO3[18] <sup>b</sup>	0.25*frag_organic[18] <sup>b</sup>
18	frag_SO3[18]		0.67*frag_SO3[64] <sup>b</sup> , 0.67*frag_SO3[48] <sup>b</sup>	1*frag_organic[44] <sup>b</sup>
19	frag_SO3[19]		0.000691*frag_SO3[18] <sup>a</sup> , 0.002*frag_SO3[17] <sup>a</sup>	0.000691*frag_organic[18] <sup>a</sup> , 0.002*frag_organic[17] <sup>a</sup>
20	frag_SO3[20]		0.002*frag_SO3[18] <sup>a</sup>	0.002*frag_organic[18] <sup>a</sup>
...				
24	frag_SO3[24], frag_H2SO4[24]	0.005*frag_H2SO4[48] <sup>b</sup>	0.005*frag_SO3[48] <sup>b</sup>	[24], -frag_sulphate[24]
25				[25]
26				[26]
27				[27]
28				
29				[29], -frag_air[29]

m/z	frag_sulphate	frag_H2SO4	frag_SO3	frag_organic
30				0.022*frag_organic[29] <sup>a</sup>
31				
32	frag_SO3[32], frag_H2SO4[32]	0.068*frag_H2SO4[81] <sup>b</sup> , 0.068*frag_H2SO4[98] <sup>b</sup>	0.21*frag_SO3[48] <sup>b</sup> , 0.21*frag_SO3[64] <sup>b</sup>	
33	frag_SO3[33], frag_H2SO4[33]	0.0079*frag_H2SO4[32] <sup>a</sup>	0.0079*frag_SO3[32] <sup>a</sup>	
34	frag_SO3[34], frag_H2SO4[34]	0.044*frag_H2SO4[32] <sup>a</sup>	0.044*frag_SO3[32] <sup>a</sup>	
...				
37				[37], -frag_chloride[37]
38				[38], -frag_chloride[38], -frag_air[38]
...				
41				[41], -frag_K[41]
42				[42]
43				[43]
44				[44], -frag_air[44]
45				[45]
...				
48	frag_SO3[48], frag_H2SO4[48]	0.465*frag_H2SO4[81] <sup>b</sup> , 0.465*frag_H2SO4[98] <sup>b</sup>	[48], -frag_organic[48], -frag_nitrate[48], -frag_H2SO4[48]	0.5*frag_organic[62]
49	frag_SO3[49], frag_H2SO4[49]	0.00829*frag_H2SO4[48] <sup>a</sup> , 0.015*frag_H2SO4[81] <sup>b</sup> , 0.015*frag_H2SO4[98] <sup>b</sup>	0.00829*frag_SO3[48] <sup>a</sup>	[49], -frag_sulphate[49]
50	frag_SO3[50], frag_H2SO4[50]	0.0462*frag_H2SO4[48] <sup>a</sup>	0.0462*frag_SO3[48] <sup>a</sup>	[50], -frag_sulphate[50]
51				[51]
52	frag_SO3[52], frag_H2SO4[52]	0.000299*frag_H2SO4[48] <sup>a</sup>	0.000299*frag_SO3[48] <sup>a</sup>	[52], -frag_sulphate[52]
53				[53]
54				[54]
55				[55]
56				[56]
57				[57]
58				[58]
59				[59]
60				[60]
61				[61]
62				[62]
63				[63], -frag_nitrate[63]
64	Frag_SO3[64], frag_H2SO4[64]	0.465*frag_H2SO4[81] <sup>b</sup> , 0.465*frag_H2SO4[98] <sup>b</sup>	[64], -frag_organic[64], -frag_H2SO4[64]	0.5*frag_organic[50] <sup>b</sup> , 0.5*frag_organic[78] <sup>b</sup>
65	Frag_SO3[65], frag_H2SO4[65]	[65], -frag_organic[65], -frag_SO3[65]	0.00868*frag_SO3[64] <sup>a</sup>	0.5*frag_organic[51] <sup>b</sup> , 0.5*frag_organic[79] <sup>b</sup>
66	Frag_SO3[66], frag_H2SO4[66]	0.0482*frag_H2SO4[64] <sup>a</sup> , 0.004*frag_H2SO4[81] <sup>b</sup> , 0.004*frag_H2SO4[98] <sup>b</sup>	0.0482*frag_SO3[64] <sup>a</sup>	[66], -frag_sulphate[66]
67				[67]
68				[68]
69				[69]
70				[70]
71				[71]
72				[72]
73				[73]
74				[74]

<i>m/z</i>	frag_sulphate	frag_H2SO4	frag_SO3	frag_organic
75				[75]
76				[76]
77				[77]
78				[78]
79				[79]
80	Frag_SO3[80], frag_H2SO4[80]	0.75*[80] <sup>b</sup> , -0.75*frag_organic[80] <sup>b</sup>	0.25*[80] <sup>b</sup> , -0.25*frag_organic[80] <sup>b</sup>	0.75*frag_organic[94] <sup>b</sup>
81	Frag_H2SO4[81]	[81], -frag_organic[81]		0.5*frag_organic[67] <sup>b</sup> , 0.5*frag_organic[95] <sup>b</sup>
82	Frag_SO3[82], frag_H2SO4[82]	0.0502*frag_H2SO4[80] <sup>a</sup> , 0.00922*frag_H2SO4[81] <sup>a</sup>	0.0502*frag_SO3[80] <sup>a</sup>	[82], -frag_sulphate[82]
83	Frag_H2SO4[83]	0.0502*frag_H2SO4[81] <sup>a</sup>		[83], -frag_sulphate[83]
84	Frag_SO3[84], frag_H2SO4[84]	.000488*frag_H2SO4[80] <sup>a</sup>	.000488*frag_SO3[80] <sup>a</sup>	[84], -frag_sulphate[84]
85	Frag_H2SO4[85]	.000488*frag_H2SO4[81] <sup>a</sup>		[85], -frag_sulphate[85]
86				[86]
87				[87]
88				[88]
89				[89]
90				[90]
91				[91]
92				[92]
93				[93]
94				[94]
95				[95]
96				[96]
97				[97]
98	Frag_H2SO4[98]	[98], -frag_organic[98]		0.5*frag_organic[84] <sup>b</sup> , 0.5*frag_organic[112] <sup>b</sup>
99	Frag_H2SO4[99]	0.00976*frag_H2SO4[98] <sup>a</sup>		[99], -frag_sulphate[99]
100	Frag_H2SO4[100]	0.0522*frag_H2SO4[98] <sup>a</sup>		[100], -frag_sulphate[100]
101				[101]
102	Frag_H2SO4[102]	0.00059*frag_H2SO4[98] <sup>a</sup>		[102], -frag_sulphate[102]

Table 5.4.III. The fragmentation tables for total sulphate (frag\_sulphate), sulphur trioxide (frag\_SO3), sulphuric acid (frag\_H2SO4) and organics (frag\_organic). When sulphate compounds are vaporised, H<sub>2</sub>SO<sub>4</sub>, SO<sub>3</sub> and H<sub>2</sub>O are produced in the detection region in varying proportions, depending on the particle composition. Note that the H<sub>2</sub>O contribution from the vaporisation of H<sub>2</sub>SO<sub>4</sub> is included in frag\_SO3, as it is formed in equal molar quantities with the SO<sub>3</sub>. The sulphate mass spectrum is taken to be the sum of these spectra. Any peaks in the ensemble mass spectrum of an *m/z* greater than 102 are assumed to be organic during normal ambient sampling. The exceptions are *m/z* 149, 180, 182, 183 and 184, as these channels tend to have high background levels in the instrument and would therefore introduce too much inherent noise to the summed signal. However, as the fraction of the total organic mass residing in the *m/z* >100 regime is typically small, these omissions do not significantly affect the overall calculation.

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## 5.5. Code availability

The source code and fragmentation tables for the analysis tools presented in this thesis are available at [http://cloudbase.phy.umist.ac.uk/people/allan/ja\\_igor.htm](http://cloudbase.phy.umist.ac.uk/people/allan/ja_igor.htm). The analysis software requires a PC or Mac running Igor Pro version 4.05 or higher. See <http://www.wavemetrics.com> for more information. 1 Gb of RAM is recommended for the larger datasets.

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## 5.6. References

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