Electronic Supplementary Material (ESI) for Faraday Discussions.

Supplement:

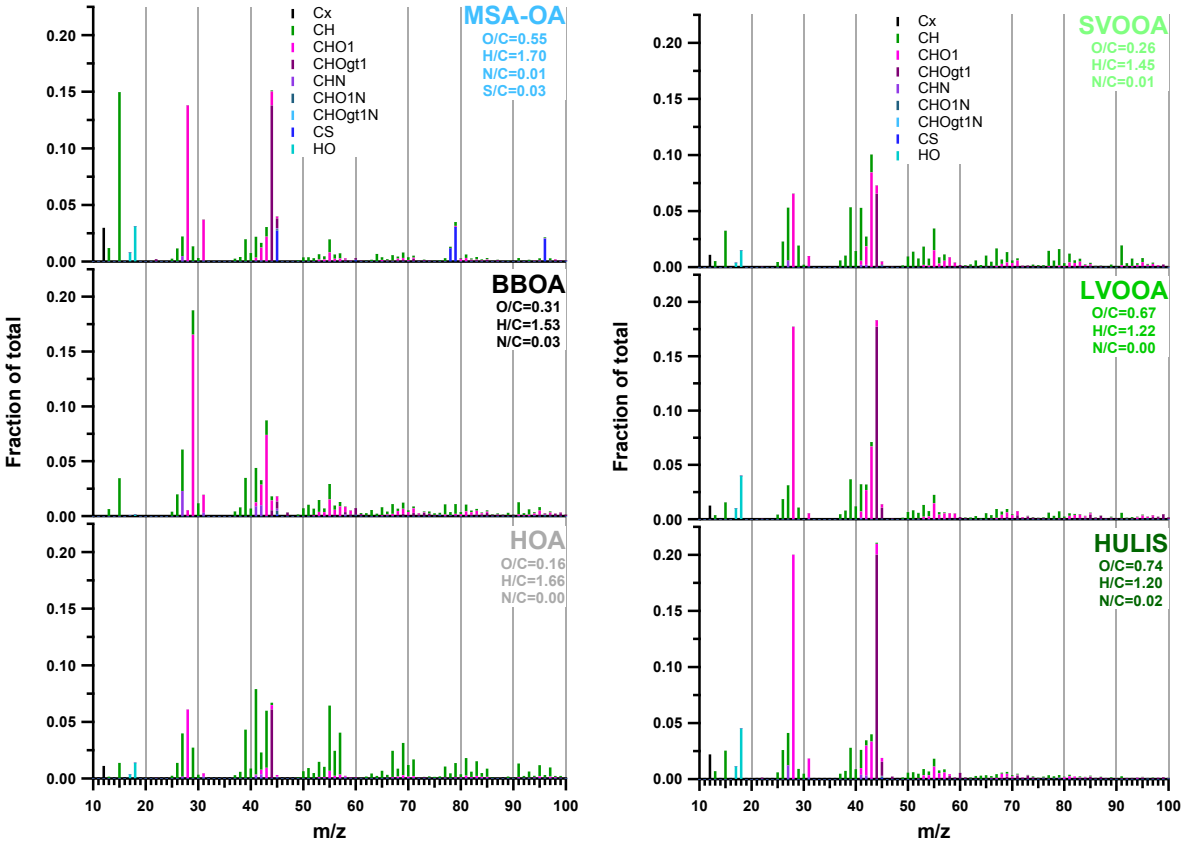


Figure S1: Mass spectra of PMF factors from AMS ground based measurements. The HR mass peaks are stacked at each unit mass m/z and colored by their chemical family (gt1 = greater than 1).

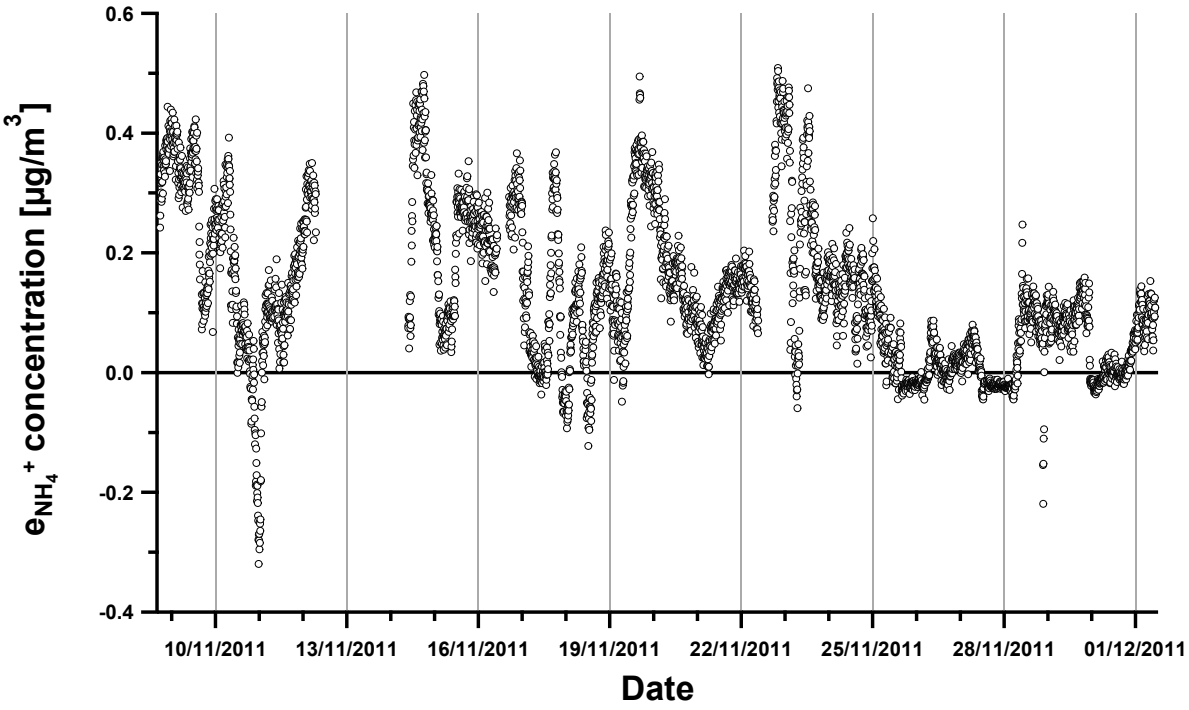


Figure S2: Time series of excess ammonium at the CESAR tower in November 2011. A campaign average of 0.13 µg/m^3 was determined, contributing 12% and 1% to total NH_4 and total aerosol mass, respectively.

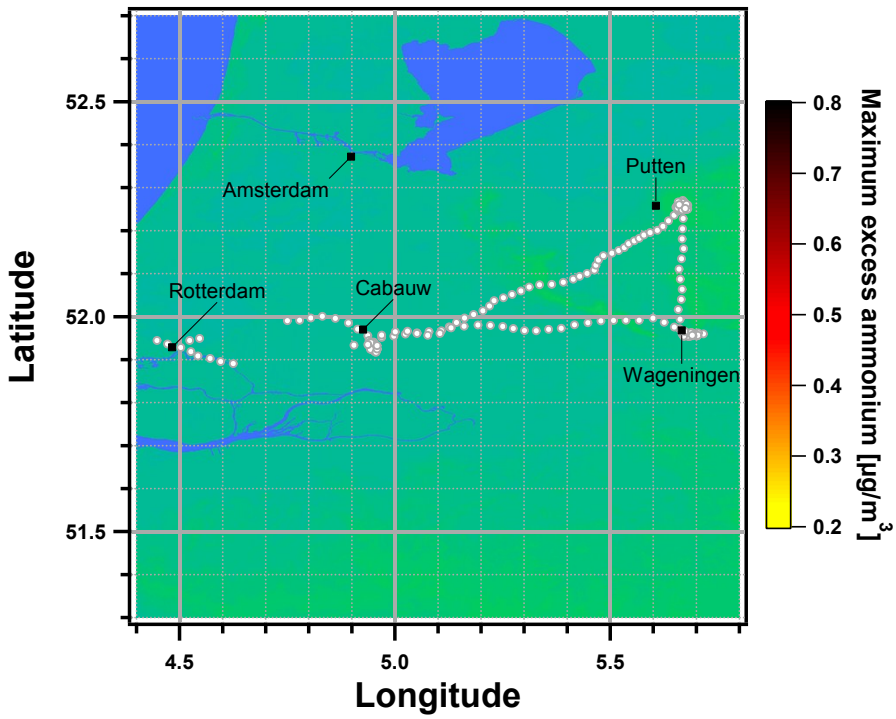
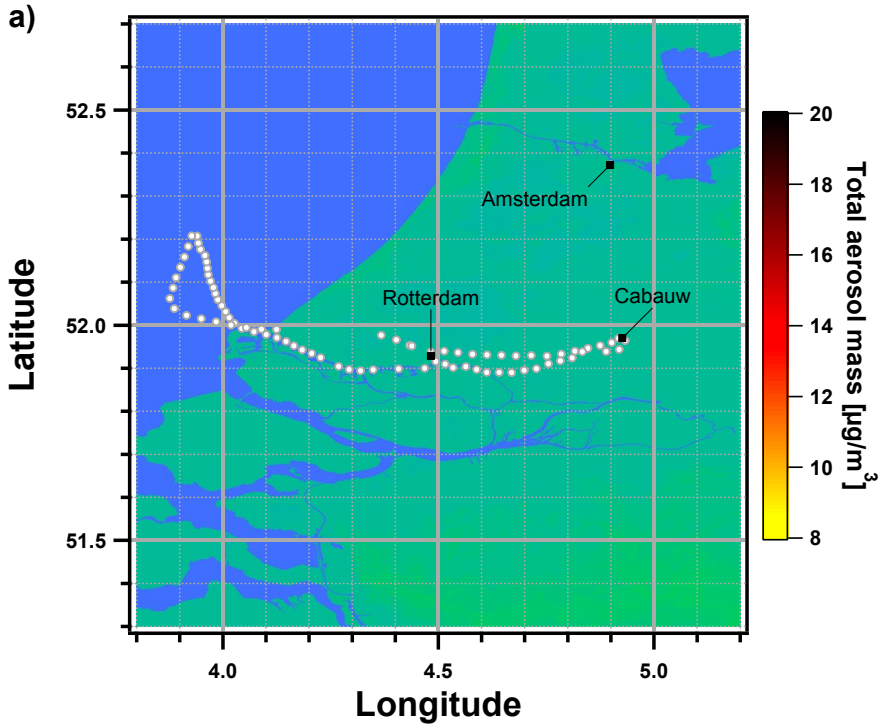


Figure S3: Zeppelin flight track on May 21st, color coded with the maximum e_{NH4} concentration



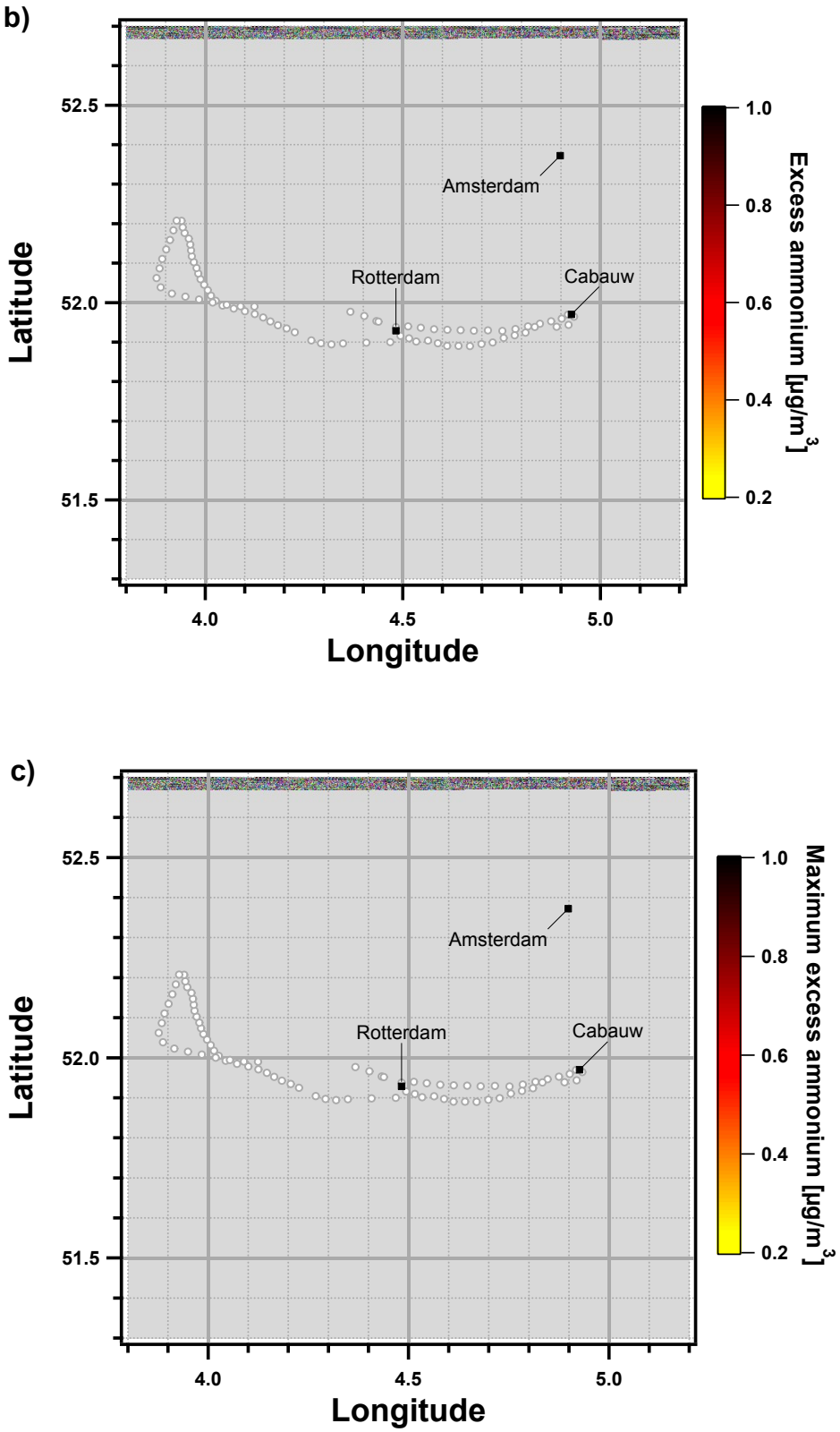


Figure S4: Zeppelin flight tracks on May 22nd, color coded with the a) total PM_1 mass concentrations, b) the e_{NH4} concentration, and c) the maximum e_{NH4} concentration

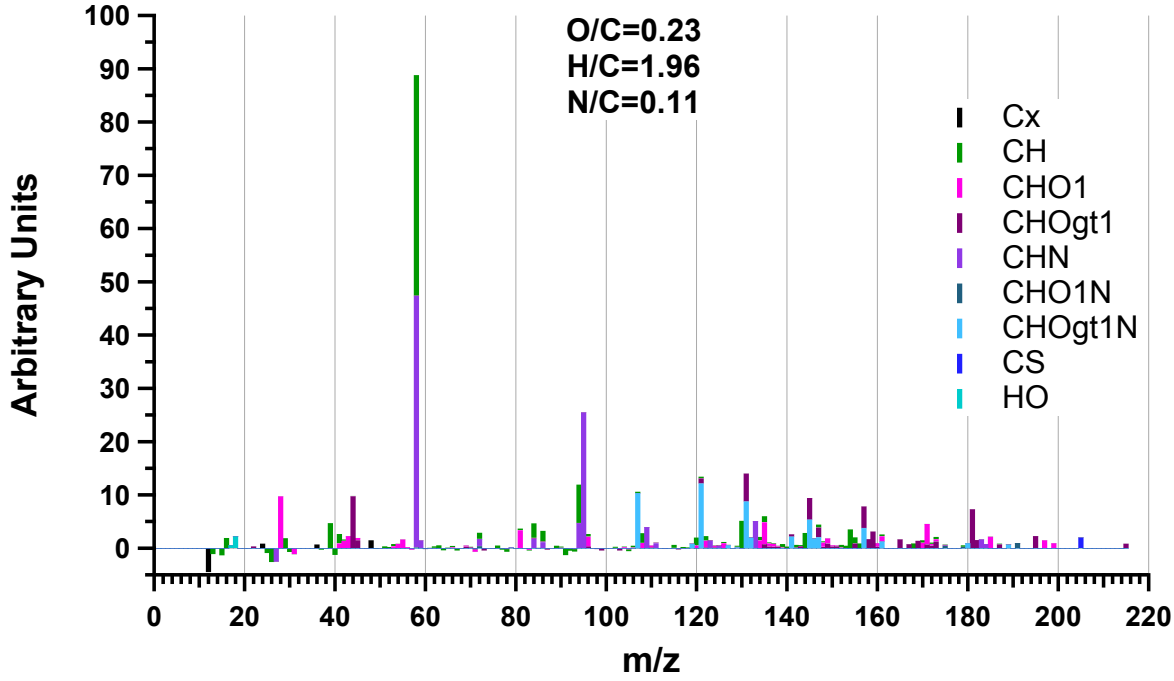


Figure S5: Mass spectrum of the residuum using the ME-2 solver on the ZAMS data set for the whole flight on May 21st. The respective residuum for the whole flight on May 22nd has a very similar pattern. The HR mass peaks are stacked at each unit mass m/z and colored by their chemical family (gt1 = greater than 1).

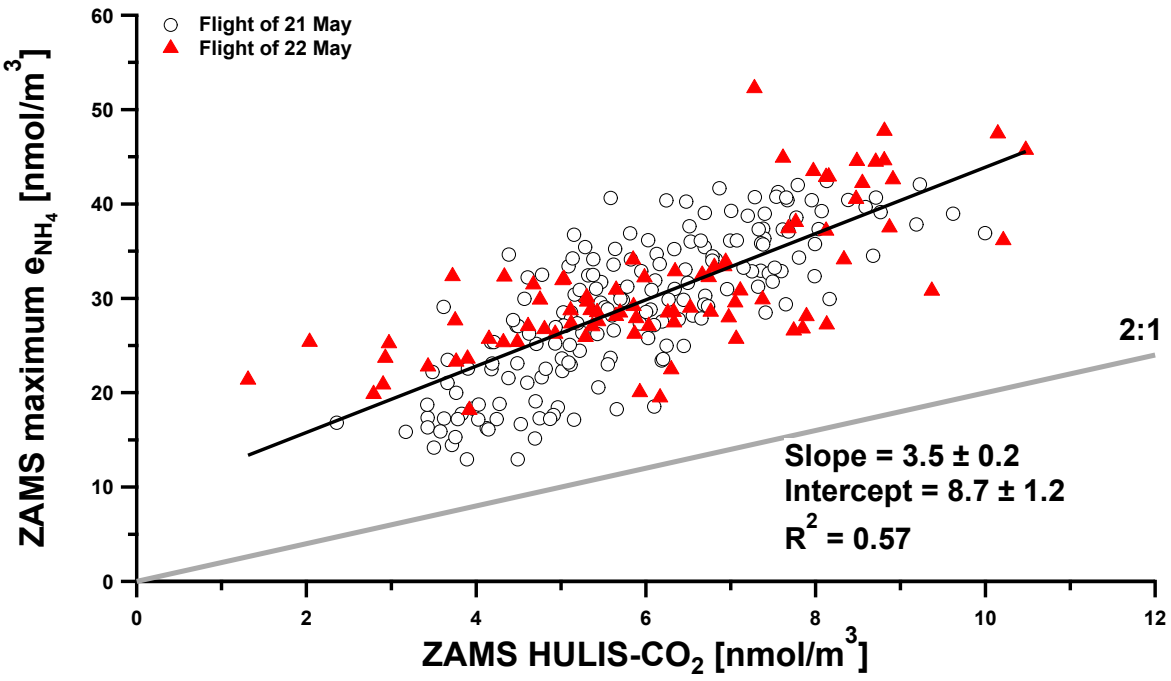


Figure S6: Correlation plots of the maximum ZAMS n_{eNH4} and $n_{HULIS\text{-}CO2}$ found for both entire Zeppelin flights combined, respectively. Note that by definition, PMF factors only consist of positive data values.

Table S1: Correlation coefficients (R^2) of the comparison between AMS PMF factor and tracer time series for the AMS ground based measurements over the entire campaign

Tracer	**HOA**	**BBOA**	**MSA-OA**	**HULIS**	**LVOOA**	**SVOOA**
NO_3	0.22	0.69	0.08	0.23	0.42	0.17
SO_4	0.17	0.43	0.16	0.30	0.36	0.02
NO_3	0.24	0.71	0.11	0.29	0.46	0.14
NH_4	0.25	0.72	0.13	0.31	0.49	0.12
Chl	0.22	0.17	0.21	0.07	0.08	0.00
eBC	0.49	0.61	0.09	0.13	0.55	0.27
Excess-NH_4	0.10	0.49	0.01	0.36	0.64	0.18
Organic-NO_3	0.10	0.26	0.00	0.17	0.29	0.47
$C_2H_4O^{2+}$ (m/z 60)	0.17	0.78	0.04	0.41	0.83	0.34
$C_3H_5O^{2+}$ (m/z 73)	0.19	0.79	0.05	0.34	0.92	0.32
CH_3SO^{2+} (m/z 79)	0.16	0.15	0.99	0.01	0.08	0.04
NO_x (gas phase)	0.60	0.21	0.06	0.02	0.12	0.16
CO (gas phase)	0.25	0.51	0.01	0.22	0.44	0.20

Table S2: Correlation coefficients (Pearson-R^2) of the comparison between excess ammonium and tracer time series for ToF-AMS (entire campaign) and ZAMS (both entire flights; number of correlated data points are written at the top of the columns) data sets, respectively. Highest important correlation coefficients are written in bold values. Note that for the correlation of the ZAMS e_{NH4} and the PMF factors a fully constrained PMF exploration was performed for the whole flight data set. The numbers in brackets represent the respective coefficients for the maximum ZAMS e_{NH4} time series.

Tracer	**e_{NH4} (ToF-AMS): 11582 points**	**e_{NH4} (ZAMS, 21st): 187 points**	**e_{NH4} (ZAMS, 22nd): 85 points**
Organics	0.62	0.75 (0.71)	0.18 (0.29)
NO_3	0.33	0.94 (0.81)	0.67 (0.88)
SO_4	0.33	0.26 (0.27)	0.50 (0.42)
NH_4	0.43	0.94 (0.63)	0.80 (0.95)
Chl	0.11	0.64 (0.54)	0.03 (0.01)
(e-)BC	0.36	0.24 (0.23)	0.00 (0.00)
Org-CO_2	**0.66**	0.77 (0.75)	0.53 (0.69)
HOA	0.10	0.04 (0.04)	0.05 (0.06)
BBOA	**0.49**	0.01 (0.01)	0.06 (0.04)
SVOOA	0.18	0.38 (0.42)	0.00 (0.00)
LVOOA	**0.64**	0.10 (0.07)	0.20 (0.28)
HULIS	0.36	**0.54 (0.61)**	**0.50 (0.52)**
MSA	0.01	0.04 (0.12)	0.19 (0.15)
Residuals	-	0.44 (0.45)	0.12 (0.19)
Rn (gas phase, 20 m height)	0.33	-	-
CO_2 (gas phase, 20 m height)	0.04	-	-
CH_4 (gas phase, 20 m height)	0.18	-	-
NO_x (gas phase)	0.12	0.10 (0.11)	0.15 (0.15)
CO (gas phase)	0.37	0.11 (0.10)	0.00 (0.00)