Supplementary information
Characterisation of aged continental aerosols over the Northern Indian Ocean

Karin Stegelius[1], Pauline Thompson[1], Pia Karbiener[1,*], Piotr Markuszewski[1,2,3], Andre S. H. Prevot[4], Sachchida N. Tripathi[5,6], Claudia Mohr[4,7], Liine Heikkinen[1,2], Krishnakant Budhavant[8,9] and Sophie L. Haslett[1,2]

[1]Department of Environmental Science, Stockholm University, Stockholm, Sweden
[2]Bolin Centre for Climate Research, Stockholm University, Stockholm, Sweden
[3]Physical Oceanography Department, Institute of Oceanology, Polish Academy of Sciences, Sopot, Poland
[4]PSI Center for Energy and Environmental Sciences, Paul Scherrer Institute, Villigen PSI, Switzerland
[5]Department of Civil Engineering, Indian Institute of Technology Kanpur, Kanpur, India
[6]Department of Sustainable Energy Engineering, Indian Institute of Technology Kanpur, Kanpur, India
[7]Department of Environmental Systems Science, ETH Zurich, Zürich, Switzerland
[8]Maldives Climate Observatory at Hanimaadhoo, Hanimaadhoo, Maldives
[9]Divecha Centre for Climate Change, Indian Institute of Science, Bangalore, India

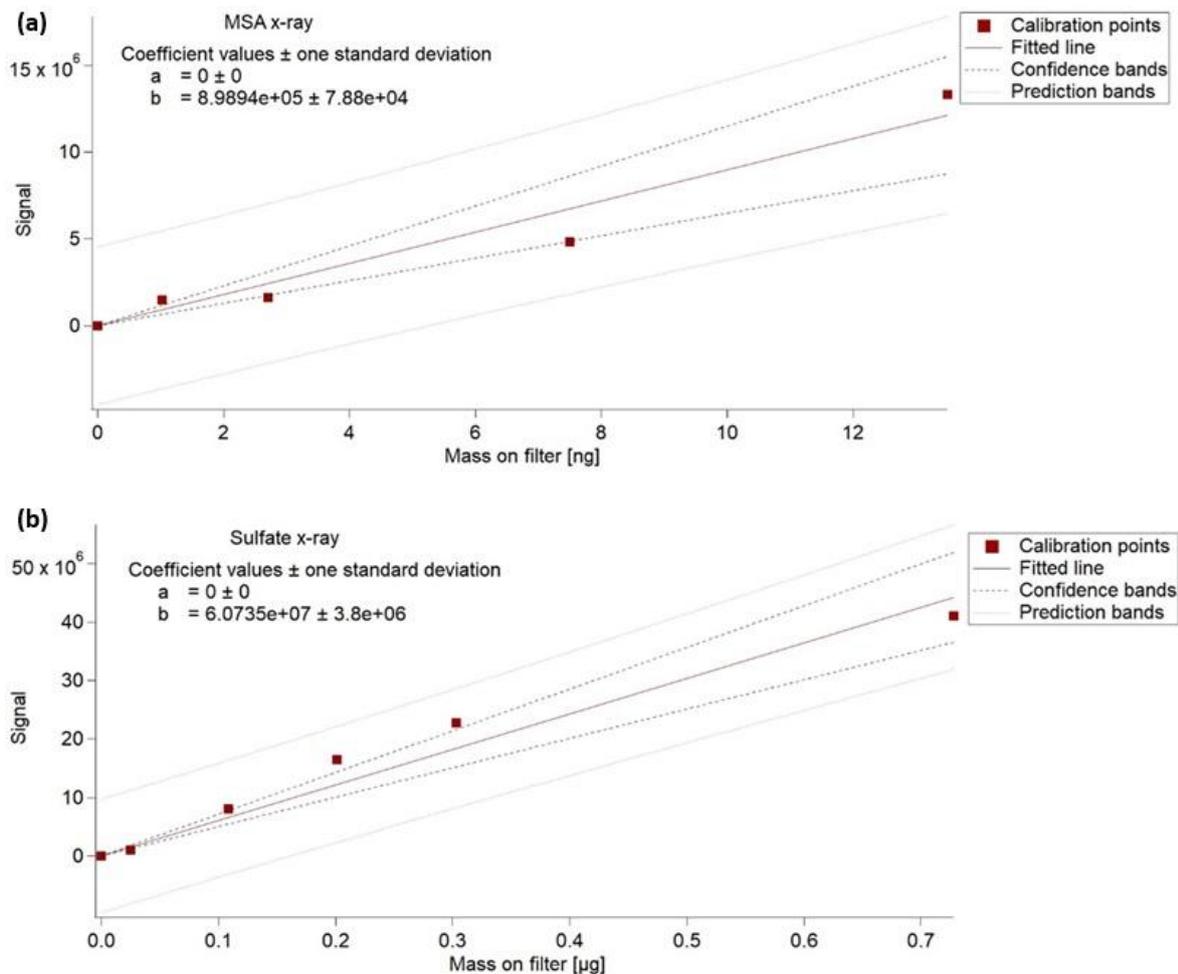*now at Department of Environmental Sciences, University of Basel, Basel, Switzerland

Figure S1: The calibration curves for (a) MSA using a x-ray source and (b) Sulfate represented by the peak of SO3-I using a x-ray source, the red squares are the calibration points, the red line the calculated straight line when forced through zero, the black dotted line are the confidence band (within which model points are expected to fall) and the light grey dotted line are prediction band (within which measured points are expected to fall) for a 95% confidence interval. In addition, the coefficient values for the fitted line are also displayed. A two-tailed t-test (alfa 0.05) showed that the intercept was non- significant for the MSA case and for the sulfate case it was a close call, why the decision was made to calculate both calibration curves with the line forced through zero.
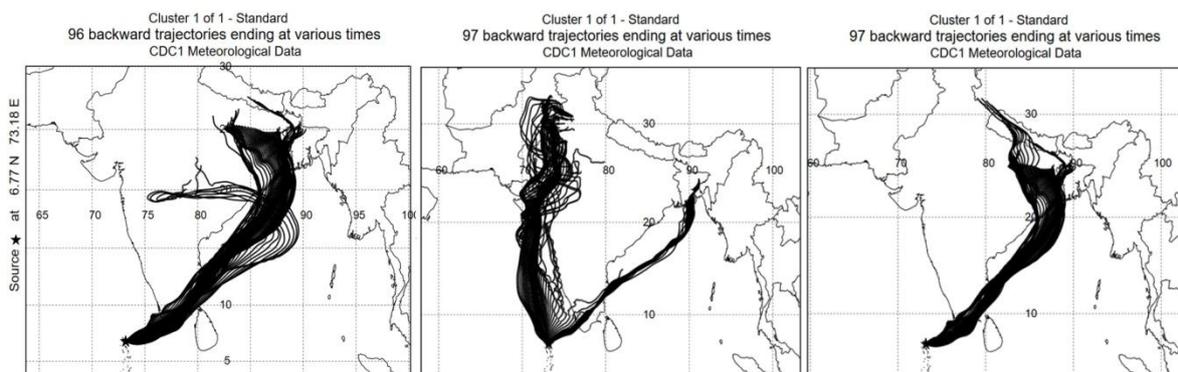

Figure S2: 10-day back trajectories generated hourly for period 1 (15–19 January 08:00–08:00(MVT)), period 2 (30 January–3 February 09:00–09:00(MVT)) and period 3 (14–18 February 06:00–06:00(MVT))
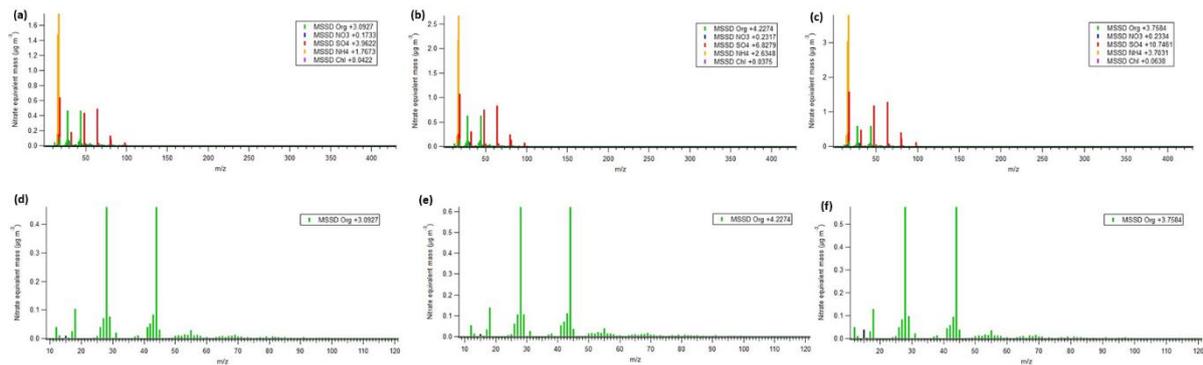
Figure S3: Mass spectra for all species observed with the AMS; Org, NO₃, SO₄, NH₄ and Chl during the three different time periods (a) period 1, (b) period 2 and (c) period 3 and the mass spectra for the organics only during (d) period 1, (e) period 2 and (f) period 3.
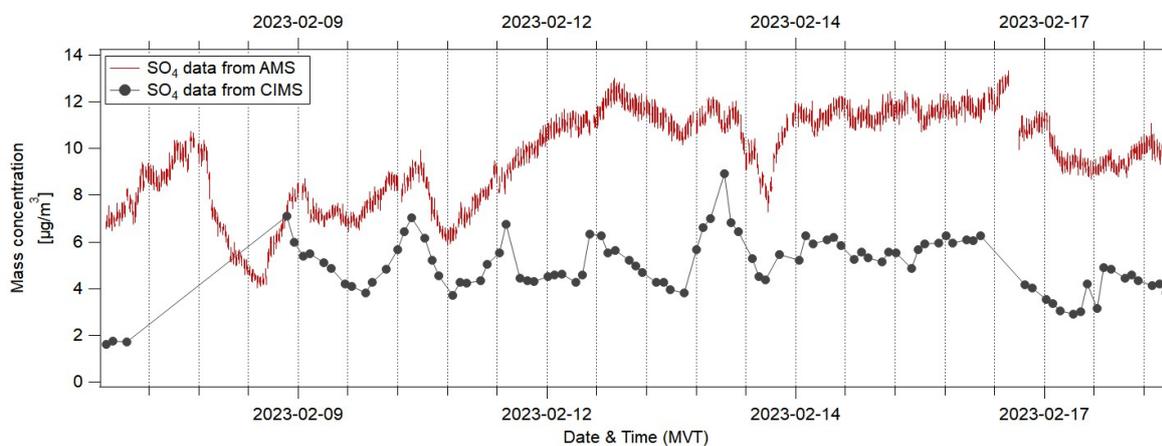

Figure S4: Time series of SO₄ data from AMS (red) and CIMS (black) during the time period 7 – 18 February 2023. The mass concentration from the CIMS is derived from calibration of the SO₃-I peak and converted from mass to a mass concentration by accounting for the flow rate and collection time at sampling, as described in section 2.4 in the main text.
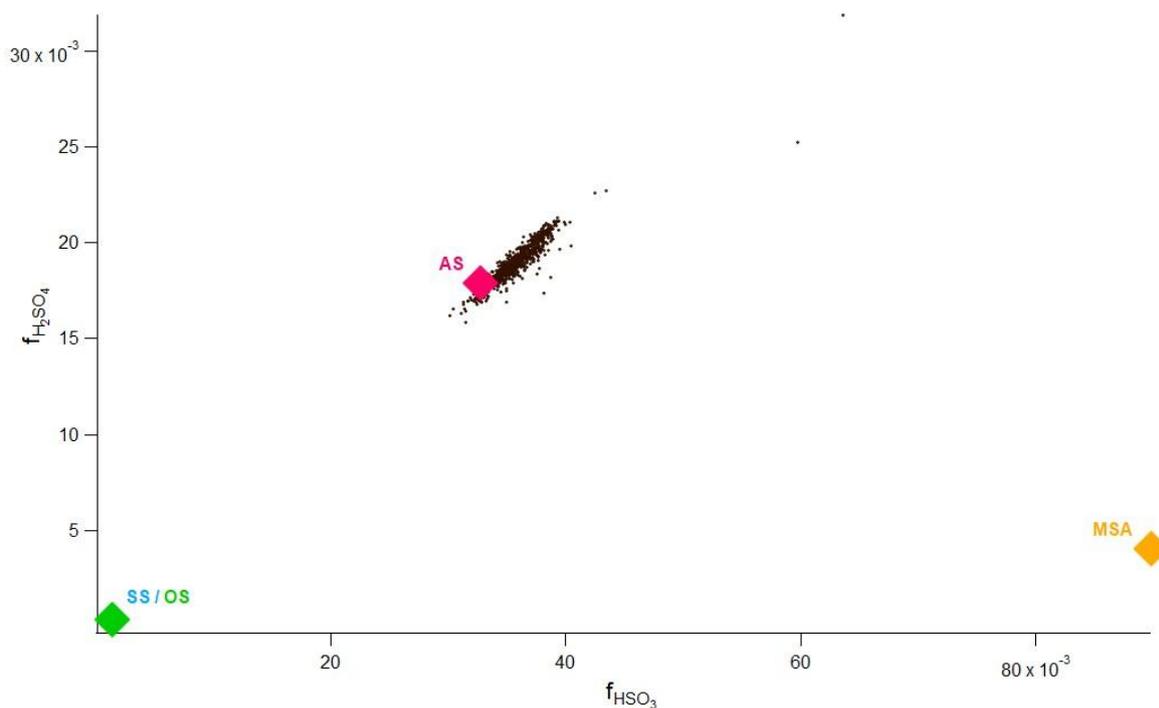
Figure S5: The fraction of H₂SO₄⁺ (f$_{H2SO4}$) and HSO₃⁺ (f$_{HSO3}$) to total SO₄ (i.e SO⁺ (m/z 48) + SO₂⁺ (m/z 64) + SO₃⁺ (m/z 80) + HSO₃⁺ (m/z 81) + H₂SO₄⁺ (m/z 98)) observed with the AMS for 4 standard compounds, Ammonium sulfate (AS), Methane sulfonic acid (MSA), Organo sulfate (OS) and Sodium sulfate (SS). The black dots show hourly averaged f$_{HSO3}$ and f$_{H2SO4}$ for all measured SO₄ during the campaign at MCOH.
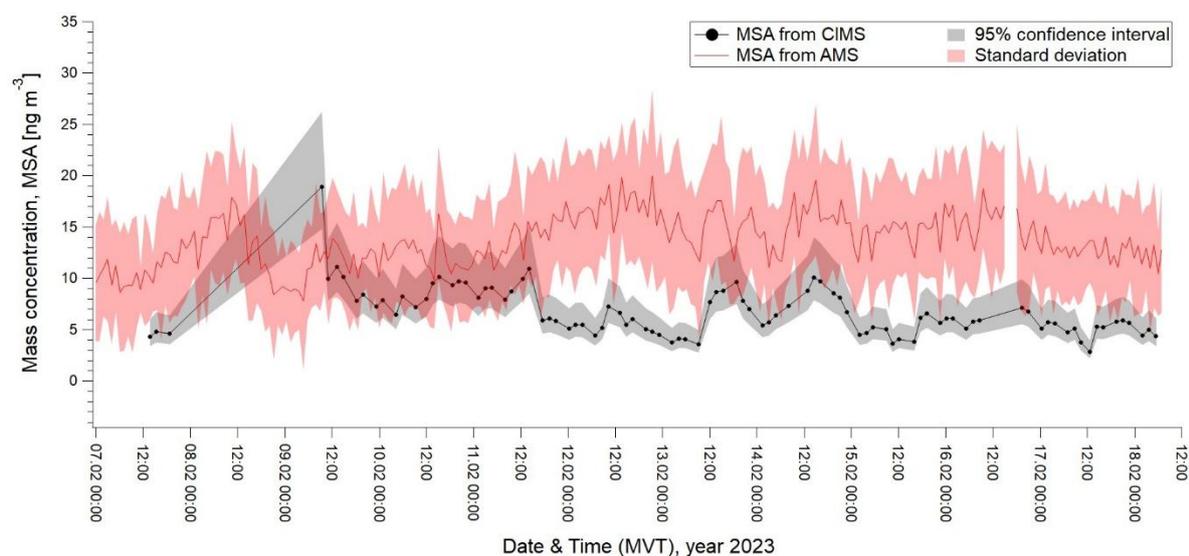

Figure S6: Time series of the MSA mass concentration (in ng m⁻³) calculated from AMS and CIMS data during the time period 7 – 18 February 2023. Data points from the AMS, shown by the red line, are hourly averaged and the standard deviation is displayed by the shaded area. Data from the CIMS are shown in black, with a shaded grey region to display the upper and lower mass concentrations based on the 95% confidence interval for the signal value in the sensitivity calibration.

Figure S7: Peak fits for the sulfur-containing organic compound signals observed by the CIMS

Text S1: Ion balance

A rough ion balance, using the approach outlined by Zhang et al. (2007), gives the following result as a campaign average:

$$\frac{NH_4(measured)}{NH_4(neutralised)} = \frac{\frac{NH_4}{18}}{2 \times \frac{SO_4}{96} + \frac{NO_3}{62} + \frac{Chl}{35.5}} = \frac{\frac{2.54}{18}}{2 \times \frac{6.49}{96} + \frac{0.181}{62} + \frac{0.0419}{35.5}} \approx \mathbf{1.01}$$

Here, *NH$_4$ (measured)* refers to measured NH$_4$ ions, while *NH$_4$ (neutralised)* refers to the ammonium (*NH$_4$*) ions that would be required to neutralise all of the observed sulfate (*SO$_4$*), nitrate (*NO$_3$*), and chloride (*Chl*) ions. A value substantially lower than 1 indicates that there is not enough NH$_4$ available to neutralise the cations, suggesting a more acidic aerosol. However, a value close to unity, as is seen here, indicates a neutral aerosol.

References

Zhang, Q., Jimenez, J. L., Worsnop, D. R., and Canagaratna, M.: A Case Study of Urban Particle Acidity and Its Influence on Secondary Organic Aerosol, Environ. Sci. Technol., 41, 3213–3219, https://doi.org/10.1021/es061812j, 2007.