

Table S1: The starting and ending date, latitude (°S), and longitude(°E) are presented for each aerosol filter deployment. The wind speed (WS; m s<sup>-1</sup>), atmospheric temperature (Atm T; °C), relative humidity (RH; %), and the number of daylight hours (hrs) were calculated as an average (Avg) over the duration of each filter deployment. For WS, Atm T and RH the standard deviations (SD) are also shown. Filter deployments are separated into early Summer (ES), Weddell Sea (WS) and late Summer (LS) depending on the location and time of sampling.

| Cruise<br>Leg ID | Collection Dates |            | Latitude (°S) |      | Longitude (°E) |       | WS (m s <sup>-1</sup> ) |     | Atm T (°C) |     | RH (%) |      | Daylight<br>(hrs) |
|------------------|------------------|------------|---------------|------|----------------|-------|-------------------------|-----|------------|-----|--------|------|-------------------|
|                  | Start Date       | End Date   | Start         | End  | Start          | End   | Avg                     | SD  | Avg        | SD  | Avg    | SD   |                   |
| ES 1             | 2018-12-07       | 2018-12-08 | 34.5          | 37.0 | 14.8           | 12.7  | 5.2                     | 1.8 | 15.7       | 0.6 | 73.4   | 4.8  | 14.5              |
| ES 2             | 2018-12-08       | 2018-12-09 | 37.1          | 41.8 | 12.6           | 8.8   | 5.0                     | 3.3 | 14.7       | 1.1 | 65.9   | 10.9 | 14.9              |
| ES 3             | 2018-12-09       | 2018-12-10 | 41.8          | 44.1 | 8.8            | 6.8   | 9.8                     | 2.3 | 10.7       | 0.7 | 100.3  | 2.7  | 15.3              |
| ES 4             | 2018-12-10       | 2018-12-12 | 45.0          | 50.8 | 6.1            | 0.6   | 13.3                    | 3.1 | 3.6        | 1.9 | 76.6   | 9.2  | 16.0              |
| ES 5             | 2018-12-14       | 2018-12-16 | 59.7          | 68.3 | 0.0            | 0.0   | 6.1                     | 2.0 | -2.6       | 0.6 | 76.1   | 8.8  | 21.3              |
| ES 6             | 2018-12-16       | 2018-12-19 | 68.5          | 70.1 | 0.0            | -2.1  | 8.8                     | 4.8 | -2.4       | 1.8 | 78.3   | 8.3  | 24.0              |
| ES 7             | 2018-12-19       | 2018-12-21 | 70.1          | 70.2 | -2.1           | -2.1  | 11.7                    | 1.8 | -3.4       | 1.9 | 69.5   | 12.3 | 24.0              |
| WS 8             | 2019-01-04       | 2019-01-06 | 67.4          | 64.0 | -16.0          | -37.0 | 8.3                     | 4.4 | -2.2       | 0.5 | 77.4   | 5.8  | 22.1              |
| WS 9             | 2019-01-06       | 2019-01-08 | 63.9          | 62.5 | -37.8          | -49.2 | 6.4                     | 3.2 | -2.0       | 0.7 | 82.9   | 7.3  | 19.8              |
| WS 10            | 2019-01-08       | 2019-01-10 | 62.5          | 65.7 | -49.2          | -60.2 | 6.9                     | 3.9 | -2.8       | 0.6 | 74.3   | 7.4  | 20.2              |
| WS 11            | 2019-01-10       | 2019-01-12 | 65.7          | 66.6 | -60.2          | -59.6 | 5.3                     | 1.4 | -2.7       | 1.0 | 69.2   | 5.1  | 21.4              |
| WS 12            | 2019-01-12       | 2019-01-14 | 66.6          | 66.1 | -59.6          | -60.4 | 4.6                     | 1.2 | -1.7       | 0.9 | 84.0   | 6.8  | 21.3              |
| WS 13            | 2019-01-14       | 2019-01-16 | 66.1          | 65.8 | -60.4          | -60.5 | 8.4                     | 2.4 | -3.9       | 1.2 | 78.7   | 4.9  | 20.6              |
| WS 14            | 2019-01-16       | 2019-01-18 | 65.8          | 65.8 | -60.5          | -60.7 | 4.6                     | 0.7 | -3.2       | 1.6 | 76.4   | 8.5  | 20.2              |
| WS 15            | 2019-01-18       | 2019-01-20 | 65.8          | 66.4 | -60.7          | -60.3 | 5.4                     | 1.1 | -3.9       | 0.6 | 83.8   | 5.3  | 20.1              |
| WS 16            | 2019-01-20       | 2019-01-22 | 66.4          | 66.4 | -60.3          | -59.9 | 3.7                     | 1.9 | -2.8       | 0.6 | 82.5   | 3.2  | 20.0              |
| WS 17            | 2019-01-22       | 2019-01-26 | 66.4          | 64.7 | -59.9          | -57.2 | 5.1                     | 2.6 | -2.9       | 0.6 | 83.4   | 5.7  | 19.2              |
| WS 18            | 2019-01-26       | 2019-01-28 | 64.7          | 63.9 | -57.2          | -52.0 | 8.2                     | 1.8 | -0.9       | 0.9 | 87.4   | 4.4  | 18.2              |
| WS 19            | 2019-01-28       | 2019-01-31 | 63.9          | 62.1 | -52.0          | -49.9 | 10.5                    | 4.8 | -1.6       | 0.9 | 87.8   | 5.0  | 17.6              |
| WS 20            | 2019-01-31       | 2019-02-01 | 62.1          | 62.2 | -50.1          | -58.9 | 7.0                     | 3.6 | -1.0       | 2.2 | 79.8   | 5.7  | 17.1              |
| WS 21            | 2019-02-01       | 2019-02-06 | 62.2          | 61.9 | -58.9          | -48.7 | 10.4                    | 3.1 | 1.5        | 0.9 | 89.4   | 5.0  | 16.8              |
| WS 22            | 2019-02-06       | 2019-02-09 | 62.0          | 69.0 | -48.2          | -52.0 | 7.7                     | 4.1 | -3.6       | 3.1 | 87.6   | 4.1  | 17.3              |
| WS 23            | 2019-02-09       | 2019-02-11 | 69.0          | 68.6 | -52.0          | -52.4 | 5.6                     | 1.8 | -6.2       | 1.8 | 80.7   | 3.3  | 17.9              |
| WS 24            | 2019-02-11       | 2019-02-13 | 68.6          | 68.8 | -52.4          | -51.8 | 3.3                     | 2.0 | -7.8       | 1.1 | 82.6   | 3.8  | 17.6              |
| WS 25            | 2019-02-13       | 2019-02-16 | 68.8          | 68.9 | -51.8          | -41.3 | 6.9                     | 3.9 | -5.2       | 1.5 | 81.7   | 6.5  | 17.2              |
| WS 26            | 2019-02-16       | 2019-02-19 | 68.9          | 69.5 | -41.3          | -8.6  | 11.9                    | 4.5 | -0.6       | 0.5 | 90.3   | 5.5  | 16.8              |
| WS 27            | 2019-02-19       | 2019-02-21 | 69.6          | 70.3 | -8.2           | -2.7  | 8.2                     | 2.9 | -5.3       | 2.3 | 76.1   | 8.1  | 16.6              |
| LS 28            | 2019-02-27       | 2019-03-01 | 69.3          | 60.0 | -4.0           | -2.3  | 8.6                     | 2.4 | -0.7       | 1.4 | 74.1   | 6.5  | 14.7              |
| LS 29            | 2019-03-01       | 2019-03-03 | 60.0          | 59.5 | -2.8           | -26.1 | 10.1                    | 4.3 | -0.2       | 0.6 | 80.8   | 5.3  | 13.9              |
| LS 30            | 2019-03-03       | 2019-03-04 | 59.5          | 55.9 | -26.1          | -33.8 | 13.0                    | 3.7 | 0.2        | 0.9 | 71.4   | 5.8  | 13.6              |
| LS 31            | 2019-03-04       | 2019-03-10 | 55.8          | 49.5 | -34.0          | 4.1   | 16.4                    | 7.5 | 4.2        | 1.4 | 88.8   | 6.6  | 13.1              |
| LS 32            | 2019-03-10       | 2019-03-12 | 49.5          | 43.1 | 4.1            | 7.8   | 11.9                    | 2.4 | 8.6        | 2.5 | 90.0   | 4.7  | 12.7              |
| LS 33            | 2019-03-12       | 2019-03-13 | 43.1          | 36.3 | 7.8            | 13.3  | 11.5                    | 1.0 | 17.5       | 2.6 | 84.3   | 4.0  | 12.5              |
| LS 34            | 2019-03-13       | 2019-03-14 | 36.1          | 34.4 | 13.4           | 17.8  | 10.2                    | 2.3 | 20.0       | 0.4 | 82.1   | 5.0  | 12.4              |

Table S2: Accepted reference values for  $\delta^{15}\text{N}$  vs.  $\text{N}_2$  in air and  $\delta^{18}\text{O}$  vs. VSMOW in ‰ for the calibration standards used. The pooled standard deviation in ‰ and sample size (1  $\text{SD}_p$ ,  $n = x$ ) is also reported.

| <b>Standard</b>       | <b>IAEA-N3</b>      | <b>USGS34</b>        | <b>USGS35</b>       | <b>Citation</b>     |
|-----------------------|---------------------|----------------------|---------------------|---------------------|
| $\delta^{15}\text{N}$ | 4.7 (0.12, n = 61)  | -1.8 (0.09, n = 61)  |                     | Böhlke et al., 2003 |
| $\delta^{18}\text{O}$ | 25.6 (0.57, n = 61) | -27.9 (0.54, n = 61) | 57.5 (0.44, n = 65) | Böhlke et al., 2003 |

Table S3: The mass weighted average N and O isotopic composition of atmospheric coarse mode  $\text{NO}_3^-$  ( $> 1\mu\text{m}$ ) are shown ( $\delta^{15}\text{N}-\text{NO}_3^-$  and  $\delta^{18}\text{O}-\text{NO}_3^-$ ; ‰), along with total coarse-mode nitrate concentration ( $[\text{NO}_3^-]$ ;  $\text{ng m}^{-3}$ ). The average (Avg) and standard deviation (SD) for the duration of each filter deployment are shown. Filter deployments are separated into early summer (ES), Weddell Sea (WS) and late summer (LS) depending on the location and time of sampling.

| Cruise<br>Leg ID | $[\text{NO}_3^-]$ ( $\text{ng m}^{-3}$ ) |      | $\delta^{15}\text{N}-\text{NO}_3^-$ (‰) |     | $\delta^{18}\text{O}-\text{NO}_3^-$ (‰) |     |
|------------------|--|------|---|-----|---|-----|
|                  | Avg                                      | SD   | Avg                                     | SD  | Avg                                     | SD  |
| ES 1             | 106.1                                    | 39.0 | -5.0                                    | 0.9 | 36.7                                    | 3.7 |
| ES 2             | 174.2                                    | 53.6 | -5.7                                    | 0.2 | 40.5                                    | 1.7 |
| ES 3             | 374.2                                    | 65.5 | -2.7                                    | 0.3 | 51.7                                    | 1.8 |
| ES 4             | 31.9                                     | 27.1 | -14.5                                   | 1.7 | 16.5                                    | 3.2 |
| ES 5             | 109.3                                    | 13.9 | -32.2                                   | 0.2 | 70.0                                    | 0.5 |
| ES 6             | 63.1                                     | 10.9 | -43.1                                   | 0.9 | 62.0                                    | 1.7 |
| ES 7             | 114.2                                    | 11.0 | -33.5                                   | 1.4 | 52.3                                    | 0.6 |
| WS 8             | 22.3                                     | 6.4  | -30.3                                   | 1.3 | 50.9                                    | 4.8 |
| WS 9             | 52.3                                     | 1.7  | -30.1                                   | 0.2 | 53.1                                    | 0.4 |
| WS 10            | 42.6                                     | 5.5  | -21.0                                   | 1.3 | 48.4                                    | 0.6 |
| WS 11            | 48.0                                     | 25.3 | -20.1                                   | 2.2 | 23.7                                    | 9.5 |
| WS 12            | 34.2                                     | 5.4  | -38.1                                   | 0.8 | 60.3                                    | 1.9 |
| WS 13            | 31.6                                     | 9.0  | -15.9                                   | 0.9 | 23.4                                    | 1.4 |
| WS 14            | 32.7                                     | 21.2 | -17.1                                   | 0.8 | 20.1                                    | 0.7 |
| WS 15            | 35.1                                     | 15.1 | -19.6                                   | 2.2 | 28.7                                    | 7.2 |
| WS 16            | 40.9                                     | 7.5  | -23.9                                   | 0.6 | 30.6                                    | 1.9 |
| WS 17            | 72.2                                     | 20.0 | -18.2                                   | 0.7 | 29.2                                    | 2.7 |
| WS 18            | 94.8                                     | 9.5  | -11.6                                   | 0.3 | 42.0                                    | 1.4 |
| WS 19            | 60.0                                     | 18.3 | -23.5                                   | 1.2 | 18.8                                    | 2.0 |
| WS 20            | 55.8                                     | 7.0  | -25.0                                   | 0.4 | 51.3                                    | 2.1 |
| WS 21            | 24.1                                     | 1.8  | -24.2                                   | 0.2 | 52.3                                    | 0.6 |
| WS 22            | 29.1                                     | 8.9  | -15.5                                   | 1.5 | 45.0                                    | 0.7 |
| WS 23            | 28.1                                     | 7.1  | -37.8                                   | 1.4 | 50.5                                    | 3.8 |
| WS 24            | 80.6                                     | 15.5 | -18.7                                   | 0.2 | 28.6                                    | 2.0 |
| WS 25            | 56.3                                     | 15.7 | -27.7                                   | 3.0 | 34.8                                    | 2.0 |
| WS 26            | 36.2                                     | 9.7  | -18.4                                   | 0.9 | 43.6                                    | 5.8 |
| WS 27            | 57.1                                     | 14.6 | -17.0                                   | 0.3 | 30.3                                    | 1.8 |
| LS 28            | 39.1                                     | 5.4  | -25.6                                   | 0.6 | 50.5                                    | 2.8 |
| LS 29            | 27.0                                     | 5.6  | -20.8                                   | 1.2 | 51.2                                    | 3.7 |
| LS 30            | 22.3                                     | 4.3  | -23.9                                   | 1.0 | 48.9                                    | 3.5 |
| LS 31            | 45.7                                     | 10.0 | -14.0                                   | 0.2 | 44.9                                    | 1.6 |
| LS 32            | 83.2                                     | 10.6 | -11.2                                   | 0.1 | 45.3                                    | 1.4 |
| LS 33            | 158.2                                    | 9.5  | -6.6                                    | 0.1 | 57.9                                    | 0.8 |
| LS 34            | 282.5                                    | 28.9 | -4.6                                    | 0.1 | 58.9                                    | 0.9 |

Table S4: Estimated contribution of alkyl nitrates, lightning NO<sub>x</sub>, and snow emissions to atmospheric NO<sub>3</sub><sup>-</sup> formation for early summer (ES) and late summer (LS) filter deployments. The δ<sup>15</sup>N signature of NO<sub>3</sub><sup>-</sup> derived from oceanic alkyl nitrates is also presented (δ<sup>15</sup>N-NO<sub>3</sub><sup>-</sup><sub>AN</sub>) for samples with an alkyl nitrate source > 10%.

| <b>Cruise leg ID</b> | <b>Alkyl nitrates (%)</b> | <b>Lightning (%)</b> | <b>Snow emissions (%)</b> | <b>δ<sup>15</sup>N-NO<sub>3</sub><sup>-</sup><sub>AN</sub> (‰)</b> |
|----------------------|---------------------------|----------------------|---------------------------|--|
| <b>ES 1</b>          | 42.7                      | 57.3                 | 0.0                       | -11.7  |
| <b>ES 2</b>          | 29.1                      | 70.8                 | 0.0                       | -19.7  |
| <b>ES 3</b>          | 4.5                       | 95.3                 | 0.0                       | -  |
| <b>ES 4</b>          | 97.0                      | 3.0                  | 0.0                       | -14.9  |
| <b>ES 5</b>          | 81.3                      | 0.0                  | 18.7                      | -28.6  |
| <b>ES 6</b>          | 35.1                      | 0.0                  | 64.9                      | -34.1  |
| <b>ES 7</b>          | 1.9                       | 0.0                  | 98.1                      | -  |
| <b>LS 28</b>         | 95.7                      | 0.0                  | 4.3                       | -24.6  |
| <b>LS 29</b>         | 89.9                      | 0.0                  | 10.1                      | -17.8  |
| <b>LS 30</b>         | 77.2                      | 0.0                  | 22.8                      | -16.8  |
| <b>LS 31</b>         | 86.4                      | 13.6                 | 0.0                       | -16.2  |
| <b>LS 32</b>         | 34.0                      | 66.0                 | 0.0                       | -33.0  |
| <b>LS 33</b>         | 26.6                      | 73.4                 | 0.0                       | -24.9  |
| <b>LS 34</b>         | 8.5                       | 91.5                 | 0.0                       | -  |