**Supplemental Material**

Cruises

Table S1. Further details of the cruises conducted as part of this study.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Cruise | Ports | | Vessel | Personnel |
| From | To |
| ANT18/1 | Bremerhaven, Germany | Cape Town, South Africa | FS *Polarstern* | A. Baker |
| JCR | Grimsby, UK | Port Stanley, Falkland Islands | RRS *James Clark Ross* | A. Baker |
| PEL | Punta Delgada, Azores | Funchal, Madeira | RV *Pelagia* | K. Timmermans |
| M55 | Curacao, Caribbean | Douala, Cameroon | FS *Meteor* | A. Baker |
| AMT12 | Port Stanley, Falkland Islands | Grimsby, UK | RRS *James Clark Ross* | T. Jickells |
| AMT13 | Immingham, UK | Port Stanley, Falkland Islands | RRS *James Clark Ross* | A. Baker |
| 24N | Freeport, Bahamas | Tenerife, Canary Islands | RRS *Discovery* | R. Mather |
| AMT14 | Port Stanley, Falkland Islands | Grimsby, UK | RRS *James Clark Ross* | K.F. Biswas |
| FEEP | Santa Cruz, Tenerife | Santa Cruz, Tenerife | FS *Poseidon* | J. Dixon |
| AMT15 | Southampton, UK | Cape Town, South Africa | RRS *Discovery* | M. Waeles |
| AIM | Las Palmas, Canaries | Mindelo, Cape Verde | FS *Poseidon* | H. Bange |
| AMT16 | Cape Town, South Africa | Falmouth, UK | RRS *Discovery* | S. Ussher |
| AMT17 | Glasgow, UK | Port Elizabeth, South Africa | RRS *Discovery* | T. Lesworth |
| ANT23/1 | Bremerhaven, Germany | Cape Town, South Africa | FS *Polarstern* | P. Croot, C. Schlosser |
| P332 | Las Palmas, Gran Canaria | Las Palmas, Gran Canaria | FS *Poseidon* | C. Powell |
| M68/3 | Santa Cruz, Tenerife | Santa Cruz, Tenerife | FS *Meteor* | M. Martino |
| P348 | Las Palmas, Gran Canaria | Las Palmas, Gran Canaria | FS *Poseidon* | H. Bange |
| RMB | Lisbon, Portugal | Falmouth, UK | RRS *Discovery* | J. Allen |
| INSPIRE | Santa Cruz, Tenerife | Santa Cruz, Tenerife | RRS *Discovery* | R. Chance |
| D326 | Santa Cruz, Tenerife | Santa Cruz, Tenerife | RRS *Discovery* | C. Powell |
| ANT24-4 | Punta Arenas, Chile | Bremerhaven, Germany | FS *Polarstern* | J. Erbland |
| AMT18 | Immingham, UK | Port Stanley, Falkland Islands | RRS *James Clark Ross* | J. Pearman |
| ICON | Santa Cruz, Tenerife | Santa Cruz, Tenerife | RRS *Discovery* | S. Thomas |
| AMT19 | Falmouth, UK | Punta Arenas, Chile | RRS *James Cook* | M. Cheize |
| P399 | Las Palmas, Gran Canaria | Lisbon, Portugal | FS *Poseidon* | H. Bange |
| AMT20 | Southampton, UK | Punta Arenas, Chile | RRS *James Cook* | I. Grefe |
| D361 | Santa Cruz, Tenerife | Santa Cruz, Tenerife | RRS *Discovery* | R. Chance |
| AMT21 | Avonmouth, UK | Punta Arenas, Chile | RRS *Discovery* | C. Yodle |

Aerosol Collection and Analysis

*Substrates*

Aerosol was collected onto Whatman 41 substrates ([Baker et al. 2007](#_ENREF_5)), with the following exceptions:

ANT18-1 - impactor stages for TM analysis were collected onto quartz filters

INSPIRE – glass fibre filters

AMT21 – MI samples were collected onto glass fibre substrates.

*Filter washing procedures*

Whatman 41 substrates to be used only for MI analysis were not washed prior to use. Glass fibre substrates for MI analysis were washed in ultrapure water (in 2 two-litre baths for ~ 1 hour, with extensive ultrapure water rinsing after each wash), dried under a laminar flow cabinet and then ashed at 450°C for 3 hours. These substrates were stored in aluminium foil before and after use.

Substrates to be used only for TM analysis were washed in 0.5M HCl and then 0.1M HNO3, with extensive rinsing with ultrapure water after each wash ([Baker et al. 2007](#_ENREF_5)).

Substrates to be used for both MI and TM analysis were washed in 0.5M HCl and then 0.1M HCl, with extensive rinsing with ultrapure water after each wash ([Rickli et al. 2010](#_ENREF_7)).

Acid-washed substrates were then dried under a laminar flow cabinet and stored in individual zip-lock plastic bags.

Exceptions to this were ANT18-1, JCR and PEL, for which the wash solutions were 0.1M HCl (x2), 0.1M HCl /1M ammonium acetate and 10% HCl /1% HNO3 respectively ([Baker et al. 2006](#_ENREF_1)).

Filter washing procedures for each cruise are summarised in Table S2.

Table S2. Filter washing procedures used for the cruises reported in this study.

|  |  |  |
| --- | --- | --- |
| Analysis Type | Washing solutions | Cruises |
| MI (Glass fibre) | Ultrapure water (x2) | INSPIRE, AMT21 |
| TM | 0.1M HCl (x2) | ANT18-1 |
| TM | 0.1M HCl, then 1M ammonium acetate | JCR |
| TM | 10% HCl, then 1% HNO3 | PEL |
| TM | 0.5M HCl, then 0.1M HNO3 | M55, AMT12, AMT13, AMT14, AMT15, AMT17, D326, AMT18, AMT19, AMT20, D361, AMT21 |
| MI/TM | 0.5M HCl, then 0.1M HCl | FEEP, AIM, AMT16, ANT23-1, P332, M68/3, P348, ICON, P399 |

*Sample collection*

Aerosol samples were collected using Tisch high volume mass flow controlled or volumetric controlled samplers operating at flow rates of ~ 1 m3 min-1. Sample collection periods were ~24 hours in almost all cases, and for most cruises sampling was interrupted manually if there was a risk of contamination of the samples from the ship. For cruises AMT20, D361 and AMT21, an automatic wind sector controller was employed to suspend sampler pump operation under flow conditions likely to result in contamination. In some cases, ship’s course coincided with wind direction for extended periods, resulting in suspension of sampling for several days (see, for example Figure 1 of [Lesworth et al. (2010)](#_ENREF_6)). For one cruise (ICON), sampling interruption procedures were not always followed resulting in contamination of some samples with ship’s stack emissions. This was evident as gross blackening of the collection substrates and 9 of the 31 samples collected during the cruise were rejected on this basis. We used soluble vanadium (as an indicator) of fuel oil combustion to confirm that the other samples collected during this cruise were not contaminated in this way.

Where cascade impactors were employed, stages 3 and 4 of the impactor were used during sampling and the substrates from these stages combined at the point of extraction to give a size split of > 1 µm (impactor stages) and < 1 µm (backup filter). For three samples, during cruises ANT18-1, D361 and AMT21, all six impactor stages were used (and analysed separately). After sampling, collection substrates were immediately sealed in separate zip-lock plastic bags and transferred to a -20°C freezer for transport to the University of East Anglia and storage before analysis.

*Extraction procedures*

Soluble aerosol species were extracted by adding 20 or 25 mL of aqueous solution to one quarter of the aerosol substrate in 50 mL polyethylene tubes, agitation for a fixed period and then filtration at 0.2 µm (Baker et al. 2007). Extraction solutions and agitation methods are summarised in Table S3. All equipment used for TM extraction was acid-washed before use and all handling of TM substrates and extraction solutions was done under a laminar flow cabinet within a trace metal clean facility.

Table S3. Solutions and sample agitation methods used to extract soluble aerosol species.

|  |  |  |
| --- | --- | --- |
| Analyte | Extraction solution | Agitation |
| NH4+ & NO3- | ultrapure water | Ultrasonication, 60 min  Mechanical shaking, 30 min (AMT21) |
| SP | 1 mM NaHCO3 | Ultrasonication, 60 min |
| Sol TMs | 1M ammonium acetate | Occasional shaking, 60 – 120 min |

*Analysis Methods*

For all cruises NO3- was determined by ion chromatography (IC) and for most cruises NH4+ was determined simultaneously using a dual-channel IC instrument. Representative chromatography conditions are given by Baker et al. (2007). For AMT19 and AMT20 NH4+ was determined using segmented flow colorimetric analysis. NH4+ was not determined for ANT18-1, ICON or P399.

SP was determined spectrophotometrically using the molybdenum blue method ([Baker et al. 2003](#_ENREF_2); [Baker et al. 2007](#_ENREF_5)).

Soluble TMs were determined using graphite furnace atomic absorption spectroscopy (GF-AAS; for ANT18-1, JCR, PEL and M55) or inductively coupled plasma – optical emission spectroscopy (ICP-OES; for all other cruises for which TMs were determined).

In all cases instruments were calibrated using standard solutions prepared in the same aqueous medium as the aerosol sample extracts. These standard solutions were prepared from high purity solids (N species and SP) or certified atomic absorption spectroscopy standards.

*Total TM analysis*

For ANT18-1, JCR and M55 separate portions of the aerosol substrates were digested using concentrated HNO3 and concentrated HF, as described in Baker et al. (2006 a & b). Total TM data for the other cruises reported were obtained using instrumental neutron activation analysis (INAA) – see Baker et al. ([2013](#_ENREF_4)).

Rain Collection

Rain samples were collected in pairs for each rain event. One set of equipment (polypropylene funnels and low density polyethylene (LDPE) bottles) was prepared for MI analysis by soaking in 10% v/v Decon 90 and then ultrapure water for at least 48 hours each. MI bottles were stored filled with ultrapure water until use. A second set of funnels and bottles was washed in 1.58M HNO3 for at least 48 hours for use in TM sampling. TM bottles were stored filled with 1.58mM HNO3 until use.

Funnels were deployed just before, or a soon as possible after, the onset of rain and recovered once rainfall had ceased. After recovery samples were frozen at -20°C for return to the University of East Anglia. For ANT18-1 and M55, portions of larger (> 30 mL) TM rain samples were filtered through 0.2 µm cellulose acetate filters immediately after collection and the filtrates were then treated in the same way as the unfiltered TM rain samples.

Aerosol Source Type Characterisation

Median fine plus coarse mode concentrations for the available aerosol samples for each source type, in each season are given in Table 2 of the main text. Tables S4 and S5 show corresponding information for the fine and coarse (respectively) modes individually. In cases where samples were collected without size-segregation, we calculated fine and coarse mode concentrations (Cf & Cc) from bulk concentrations (Cb) for each sample using our previously published ([Baker et al. 2010](#_ENREF_3); [Baker et al. 2013](#_ENREF_4)) median observed fractions of each species (x) in coarse mode aerosol (fc; Eqns S1 and S2).

Cxf = Cxb (1 – fc,x) (S1)

Cxc = Cxb fc,x (S1)

Calculated fine and coarse mode concentrations were then combined with observed concentrations to calculate median concentrations for each species and air mass type.

In Table S6 (a-d) we list the concentration ranges and numbers of observations used to derive the median fine plus coarse mode concentrations.

Uncertainty in Dry Deposition Flux Calculation

Uncertainties in dry deposition estimates were assessed as described in the main text. Table S7 shows, for each species, region and season, the baseline dry flux estimate and the range of fluxes calculated using estimates based on the number of samples collected and the interquartile range of the observed aerosol concentrations. Figure S1 summarises the overall uncertainty for the dry input of each species to the whole of our study region, normalised to that species’ baseline input.



**Figure S1.** Uncertainties in estimates of dry atmospheric inputs to the ETNA normalised to the baseline calculation of input to the region for each species (Table 7). Uncertainties estimated using number of samples collected for each season in the North and South are shown as black bars, while those estimated from the interquartile range for each characteristic concentration are shown as red bars. The total number of aerosol samples analysed for each species (n) is also shown.

Table S4. Median fine aerosol concentrations (nmol m-3) of NO3-, NH4+, SP and soluble (s-) and total (t-) TMs in air from different origins collected in the North and South regions of the ETNA. Air mass origin codes are defined in the main text.

|  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| *Season* Region | Air Mass | Subst a | NH4+ | NO3- | SP | s-Fe | s-Al | s-Mn | t-Fe | t-Al | t-Mn |
| *MAM* |  |  |  |  |  |  |  |  |  |  |  |
| N | NAM | S1 |  |  |  |  |  |  |  |  |  |
|  | EUR | S2 | 27.5 | 3.5 | 0.017 | 0.046 | 0.14 | 0.007 | 0.73 | 1.71 | 0.016 |
|  | RNA |  | 6.2 | 1.3 | 0.012 | 0.031 | 0.15 | 0.004 | 0.53 | 0.81 | 0.008 |
|  | SAH |  | 16.2 | 3.4 | 0.036 | 0.103 | 0.67 | 0.022 | 3.25 | 11.4 | 0.049 |
| S | RNA | S3 | 1.0 | 0.2 |  | 0.007 | 0.04 | 0.002 | 0.36 | 0.87 | 0.009 |
|  | RSA | S4 | 3.3 | 0.4 | 0.007 | 0.020 | 0.11 | 0.0002 | 0.92 | 2.15 | 0.010 |
|  | SAB | S5 |  |  |  |  |  |  |  |  |  |
|  | SAF | S2 | 5.2 | 0.7 | 0.019 | 0.023 | 0.23 | 0.008 | 0.98 | 3.37 | 0.015 |
|  | SAH |  | 11.6 | 1.3 | 0.038 | 0.045 | 0.33 | 0.014 | 1.95 | 6.82 | 0.034 |
| *JJA* |  |  |  |  |  |  |  |  |  |  |  |
| N | NAM | S1 |  |  |  |  |  |  |  |  |  |
|  | EUR | S2 |  | 3.4 |  | 0.030 | 0.09 | 0.003 |  |  |  |
|  | RNA |  | 14.5 | 1.5 | 0.020 | 0.034 | 0.17 | 0.006 | 1.45 | 4.9 | 0.028 |
|  | SAH |  | 37.4 | 3.6 | 0.060 | 0.100 | 0.48 | 0.022 | 6.31 | 21.2 | 0.090 |
| S | RNA | S3 | 19.7 | 0.5 | 0.011 | 0.015 | 0.04 | 0.002 | 0.4 | 0.9 | 0.006 |
|  | RSA | S4 |  |  |  |  |  |  |  |  |  |
|  | SAB | S5 |  |  |  |  |  |  |  |  |  |
|  | SAF | S2 | 7.4 | 1.1 | 0.018 | 0.004 | 0.03 | 0.001 | 0.56 | 2.16 | 0.011 |
|  | SAH |  | 0.7 | 2.3 | 0.010 | 0.045 | 0.42 | 0.022 | 4.15 | 14.1 | 0.063 |
| *SON* |  |  |  |  |  |  |  |  |  |  |  |
| N | NAM | S1 |  |  |  |  |  |  |  |  |  |
|  | EUR | S2 | 26.7 | 4.8 |  | 0.005 | 0.04 | 0.001 |  |  |  |
|  | RNA |  | 3.4 | 1.1 | 0.007 | 0.005 | 0.04 | 0.001 | 0.12 | 0.24 | 0.003 |
|  | SAH |  | 12.6 | 1.9 | 0.035 | 0.073 | 0.65 | 0.026 | 2.43 | 12.5 | 0.051 |
| S | RNA | S3 |  |  |  |  |  |  |  |  |  |
|  | RSA | S4 |  |  |  |  |  |  |  |  |  |
|  | SAB |  | 7.7 | 0.5 | 0.017 | 0.011 | 0.08 | 0.002 | 0.23 | 0.64 | 0.004 |
|  | SAF |  | 9.1 | 0.7 | 0.011 | 0.016 | 0.08 | 0.002 | 0.24 | 0.87 | 0.005 |
|  | SAH |  | 10.0 | 1.7 | 0.027 | 0.078 | 0.78 | 0.032 | 4.04 | 15.4 | 0.068 |
| *DJF* |  |  |  |  |  |  |  |  |  |  |  |
| N | NAM | S1 |  |  |  |  |  |  |  |  |  |
|  | EUR | S2 | 3.7 | 2.7 | 0.027 | 0.033 | 0.055 | 0.006 | 0.80 | 0.38 | 0.005 |
|  | RNA |  | 2.2 | 1.3 | 0.017 | 0.009 | 0.066 | 0.002 | 0.21 | 0.36 | 0.005 |
|  | SAH |  | 3.0 | 2.1 | 0.034 | 0.244 | 1.84 | 0.096 | 19.1 | 59.5 | 0.266 |
| S | RNA | S3 | 0.8 | 0.7 | 0.007 | 0.012 | 0.072 | 0.002 | 0.35 | 0.96 | 0.005 |
|  | RSA | S4 |  |  |  |  |  |  |  |  |  |
|  | SAB | S5 |  |  |  |  |  |  |  |  |  |
|  | SAF | S2 |  |  |  |  |  |  |  |  |  |
|  | SAH |  | 7.1 | 3.3 | 0.037 | 0.154 | 1.27 | 0.101 | 14.1 | 54.2 | 0.220 |

a – Substitutions:

S1 – used concentration for RNA type in same region, same season

S2 – used median concentration of all values available for the type

S3 – used concentration for RNA type in North region, same season

S4 – used concentration for RSA type from Region 4 (Southeast Atlantic) from Baker et al., 2010; 2013

S5 – used concentration for this type from SON.

Table S5. Median coarse aerosol concentrations (nmol m-3) of NO3-, NH4+, SP and soluble (s-) and total (t-) TMs in air from different origins collected in the North and South regions of the ETNA. Air mass origin codes are defined in the main text.

|  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| *Season* Region | Air Mass | Subst a | NH4+ | NO3- | SP | s-Fe | s-Al | s-Mn | t-Fe | t-Al | t-Mn |
| *MAM* |  |  |  |  |  |  |  |  |  |  |  |
| N | NAM | S1 |  |  |  |  |  |  |  |  |  |
|  | EUR | S2 | 4.0 | 45.2 | 0.012 | 0.030 | 0.15 | 0.010 | 0.83 | 2.46 | 0.018 |
|  | RNA |  | 1.3 | 14.7 | 0.010 | 0.027 | 0.17 | 0.008 | 0.60 | 1.16 | 0.009 |
|  | SAH |  | 0.6 | 28.5 | 0.034 | 0.114 | 0.83 | 0.063 | 6.00 | 22.6 | 0.111 |
| S | RNA | S3 | 0.6 | 2.5 |  | 0.010 | 0.21 | 0.014 | 0.40 | 1.25 | 0.010 |
|  | RSA | S4 | 1.0 | 3.7 | 0.007 | 0.015 | 0. | 0.011 | 0.54 | 2.07 | 0.017 |
|  | SAB | S5 |  |  |  |  |  |  |  |  |  |
|  | SAF | S2 | 5.3 | 8.1 | 0.027 | 0.052 | 0.45 | 0.033 | 1.99 | 6.54 | 0.037 |
|  | SAH |  | 14.7 | 18.2 | 0.038 | 0.051 | 0.37 | 0.041 | 3.59 | 13.6 | 0.077 |
| *JJA* |  |  |  |  |  |  |  |  |  |  |  |
| N | NAM | S1 |  |  |  |  |  |  |  |  |  |
|  | EUR | S2 |  | 30.4 |  | 0.022 | 0.13 | 0.006 |  |  |  |
|  | RNA |  | 2.0 | 13.3 | 0.014 | 0.041 | 0.32 | 0.014 | 1.64 | 7.02 | 0.032 |
|  | SAH |  | 3.9 | 24.3 | 0.045 | 0.134 | 1.04 | 0.071 | 11.6 | 42.2 | 0.204 |
| S | RNA | S3 | 0.6 | 8.2 | 0.007 | 0.012 | 0.07 | 0.003 | 0.4 | 1.34 | 0.007 |
|  | RSA | S4 |  |  |  |  |  |  |  |  |  |
|  | SAB | S5 |  |  |  |  |  |  |  |  |  |
|  | SAF | S2 | 1.5 | 12.9 | 0.022 | 0.004 | 0.03 | 0.003 | 1.14 | 4.19 | 0.027 |
|  | SAH |  | 0.1 | 18.3 | 0.009 | 0.033 | 0.29 | 0.042 | 7.65 | 28.1 | 0.143 |
| *SON* |  |  |  |  |  |  |  |  |  |  |  |
| N | NAM | S1 |  |  |  |  |  |  |  |  |  |
|  | EUR | S2 | 17.6 | 7.0 |  | 0.008 | 0.03 | 0.001 |  |  |  |
|  | RNA |  | 0.8 | 8.8 | 0.003 | 0.006 | 0.03 | 0.001 | 0.14 | 0.31 | 0.004 |
|  | SAH |  | 0.7 | 17.9 | 0.027 | 0.090 | 0.73 | 0.084 | 7.49 | 25.0 | 0.12 |
| S | RNA | S3 |  |  |  |  |  |  |  |  |  |
|  | RSA | S4 |  |  |  |  |  |  |  |  |  |
|  | SAB |  | 1.1 | 9.7 | 0.021 | 0.012 | 0.09 | 0.007 | 0.52 | 1.99 | 0.013 |
|  | SAF |  | 1.7 | 8.3 | 0.013 | 0.012 | 0.10 | 0.007 | 0.55 | 1.26 | 0.012 |
|  | SAH |  | 1.0 | 14.5 | 0.026 | 0.064 | 0.62 | 0.077 | 8.47 | 30.8 | 0.116 |
| *DJF* |  |  |  |  |  |  |  |  |  |  |  |
| N | NAM | S1 |  |  |  |  |  |  |  |  |  |
|  | EUR | S2 | 5.3 | 19.1 | 0.012 | 0.014 | 0.03 | 0.005 | 0.74 | 0.40 | 0.014 |
|  | RNA |  | 3.5 | 5.9 | 0.012 | 0.012 | 0.06 | 0.004 | 0.24 | 0.52 | 0.005 |
|  | SAH |  | 0.2 | 14.1 | 0.034 | 0.284 | 2.89 | 0.247 | 35.4 | 118 | 0.601 |
| S | RNA | S3 | 1.3 | 4.5 | 0.002 | 0.015 | 0.07 | 0.002 | 0.40 | 1.38 | 0.006 |
|  | RSA | S4 |  |  |  |  |  |  |  |  |  |
|  | SAB | S5 |  |  |  |  |  |  |  |  |  |
|  | SAF | S2 |  |  |  |  |  |  |  |  |  |
|  | SAH |  | 1.8 | 26.3 | 0.077 | 0.239 | 1.94 | 0.266 | 26.0 | 108 | 0.498 |

a – Substitutions:

S1 – used concentration for RNA type in same region, same season

S2 – used median concentration of all values available for the type

S3 – used concentration for RNA type in North region, same season

S4 – used concentration for RSA type from Region 4 (Southeast Atlantic) from Baker et al., 2010; 2013

S5 – used concentration for this type from SON.

Table S6a. Ranges of fine plus coarse aerosol concentrations (nmol m-3) of NO3-, NH4+, SP and soluble (s-) and total (t-) TMs in air from different origins collected in the North and South regions of the ETNA in MAM. Numbers of observations in each category are given in parentheses. Air mass origin codes are defined in the text.

|  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| *Season* Region | Air Mass | NH4+ | NO3- | SP | s-Fe | s-Al | s-Mn | t-Fe | t-Al | t-Mn |
| *MAM* |  |  |  |  |  |  |  |  |  |  |
| N | NAM | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) |
|  | EUR | 25.6-37.3  (2) | 42.0-55.5  (2) | 0.026-0.032 (2) | 0.076  (1) | 0.29  (1) | 0.017  (1) | 1.56  (1) | 4.17  (1) | 0.034  (1) |
|  | RNA | 1.6-30.9  (38) | 3.1-65.9  (50) | 0.002-0.052 (30) | 0.004-0.453  (34) | 0.04-2.95  (34) | 0.001-0.091  (34) | 0.40-3.55  (22) | 0.69-12.3  (22) | 0.004-0.056  (22) |
|  | SAH | 3.9-35.0  (12) | 14.1-93.4  (22) | 0.044-0.103 (7) | 0.076-0.550  (17) | 0.56-5.66  (17) | 0.017-0.373  (17) | 3.44-54.3  (7) | 6.51-206  (7) | 0.038-0.926  (7) |
| S | RNA | 1.1  (1) | 2.5  (1) | (0) | 0.012  (1) | 0.24  (1) | 0.016  (1) | 0.76  (1) | 2.12  (1) | 0.018  (1) |
|  | RSA | 2.0-11.9  (3) | 2.7-7.4  (3) | 0.004-0.022  (3) | 0.035  (1) | 0.23  (1) | 0.011  (1) | 1.46  (1) | 4.22  (1) | 0.027  (1) |
|  | SAB | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) |
|  | SAF | 3.6-29.5  (4) | 4.3-9.1  (4) | 0.027  (1) | 0.009-0.114  (4) | 0.09-0.78  (4) | 0.007-0.096  (4) | 0.46-7.26  (4) | 0.63-28.4  (4) | 0.007-0.160  (4) |
|  | SAH | 4.1-37.4  (13) | 11.2-43.0  (13) | 0.010-0.096  (9) | 0.043-0.212  (9) | 0.33-1.58  (9) | 0.025-0.172  (9) | 2.39-12.3  (9) | 8.09-50.6  (9) | 0.049-0.274  (9) |

Table S6b. Ranges of fine plus coarse aerosol concentrations (nmol m-3) of NO3-, NH4+, SP and soluble (s-) and total (t-) TMs in air from different origins collected in the North and South regions of the ETNA in JJA. Numbers of observations in each category are given in parentheses. Air mass origin codes are defined in the text.

|  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| *Season* Region | Air Mass | NH4+ | NO3- | SP | s-Fe | s-Al | s-Mn | t-Fe | t-Al | t-Mn |
| *JJA* |  |  |  |  |  |  |  |  |  |  |
| N | NAM | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) |
|  | EUR | (0) | 33.8  (1) | (0) | 0.052  (1) | 0.22  (1) | 0.009  (1) | (0) | (0) | (0) |
|  | RNA | 4.6-28.1  (16) | 2.1-36.6  (21) | 0.013-0.108  (12) | 0.019-0.161  (13) | 0.03-1.22  (14) | 0.006-0.106  (14) | 0.65-9.08  (9) | 2.22-20.3  (9) | 0.017-0.100  (9) |
|  | SAH | 21.8-92.3  (17) | 17.8-59.8  (24) | 0.061-0.182  (16) | 0.073-1.82  (23) | 0.44-10.1  (23) | 0.013-0.447  (23) | 6.64-88.5  (16) | 10.1-302  (16) | 0.114-1.34  (16) |
| S | RNA | 18.0-22.4  (2) | 8.1-9.2  (2) | 0.014-0.023  (2) | 0.027  (1) | 0.11  (1) | 0.005  (1) | 0.83  (1) | 2.27  (1) | 0.013  (1) |
|  | RSA | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) |
|  | SAB | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) |
|  | SAF | 7.4-10.5  (2) | 13.8-14.1  (2) | 0.032-0.048  (2) | 0.001-0.017  (2) | 0.004-0.11  (2) | 0.0001-0.008  (2) | 0.44-2.97  (2) | 1.39-11.3  (2) | 0.014-0.063  (2) |
|  | SAH | 0.8-73.5  (3) | 9.6-38.2  (3) | 0.012-0.089  (3) | 0.053-0.104  (3) | 0.39-1.37  (3) | 0.035-0.317  (3) | 8.49-44.1  (3) | 26.2-158  (3) | 0.132-0.784  (3) |

Table S6c. Ranges of fine plus coarse aerosol concentrations (nmol m-3) of NO3-, NH4+, SP and soluble (s-) and total (t-) TMs in air from different origins collected in the North and South regions of the ETNA in SON. Numbers of observations in each category are given in parentheses. Air mass origin codes are defined in the text.

|  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| *Season* Region | Air Mass | NH4+ | NO3- | SP | s-Fe | s-Al | s-Mn | t-Fe | t-Al | t-Mn |
| *SON* |  |  |  |  |  |  |  |  |  |  |
| N | NAM | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) |
|  | EUR | 44.3  (1) | 11.8  (1) | (0) | 0.013  (1) | 0.06  (1) | 0.001  (1) | (0) | (0) | (0) |
|  | RNA | 1.2-42.7  (31) | 3.4-25.4  (31) | 0.004-0.030  (11) | 0.001-0.039  (28) | 0.01-0.23  (28) | 0.0003-0.027  (28) | 0.02-1.02  (20) | 0.06-3.69  (20) | 0.002-0.023  (20) |
|  | SAH | 0.1-83.4  (33) | 5.3-63.7  (35) | 0.027-0.132  (18) | 0.040-0.775  (30) | 0.09-7.77  (30) | 0.016-0.587  (30) | 2.07-97.0  (17) | 3.54-300  (17) | 0.028-1.36  (17) |
| S | RNA | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) |
|  | RSA | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) |
|  | SAB | 0.3-19.4  (16) | 6.7-20.0  (16) | 0.013-0.076  (10) | 0.009-0.197  (16) | 0.05-1.54  (16) | 0.002-0.066  (16) | 0.36-6.53  (8) | 1.06-19.2  (8) | 0.010-0.104  (8) |
|  | SAF | 0.1-27.1  (20) | 1.9-24.6  (21) | 0.005-0.050  (15) | 0.005-0.188  (22) | 0.02-1.65  (22) | 0.002-0.062  (22) | 0.20-3.05  (15) | 1.39-10.7  (15) | 0.007-0.055  (15) |
|  | SAH | 0.1-37.8  (27) | 3.8-59.6  (28) | 0.023-0.103  (21) | 0.007-0.593  (27) | 0.11-5.71  (27) | 0.005-0.616  (27) | 2.21-66.2  (18) | 7.16-271  (18) | 0.037-1.18  (18) |

Table S6d. Ranges of fine plus coarse aerosol concentrations (nmol m-3) of NO3-, NH4+, SP and soluble (s-) and total (t-) TMs in air from different origins collected in the North and South regions of the ETNA in DJF. Numbers of observations in each category are given in parentheses. Air mass origin codes are defined in the text.

|  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| *Season* Region | Air Mass | NH4+ | NO3- | SP | s-Fe | s-Al | s-Mn | t-Fe | t-Al | t-Mn |
| *DJF* |  |  |  |  |  |  |  |  |  |  |
| N | NAM | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) |
|  | EUR | 8.0-10.1  (2) | 19.1-24.4  (2) | 0.019-0.048  (3) | 0.028-0.068  (3) | 0.08-0.44  (3) | 0.009-0.018  (3) | 0.03-2.61  (3) | 0.19-9.46  (3) | 0.004-0.052  (3) |
|  | RNA | 2.9-16.0  (18) | 1.8-18.0  (18) | 0.007-0.048  (8) | 0.007-0.101  (18) | 0.03-0.49  (18) | 0.002-0.026  (18) | 0.03-4.48  (18) | 0.24-12.5  (18) | 0.004-0.059  (18) |
|  | SAH | 0.3-38.6  (29) | 4.1-40.5  (31) | 0.020-0.405  (25) | 0.047-1.94  (28) | 0.36-15.1  (28) | 0.035-1.44  (28) | 7.11-299  (28) | 17.0-1120  (28) | 0.099-5.13  (28) |
| S | RNA | 0.2-12.9  (4) | 2.3-25.8  (4) | 0.009-0.061  (4) | 0.026-0.075  (5) | 0.13-0.45  (5) | 0.004-0.045  (5) | 0.64-1.41  (5) | 1.87-2.74  (5) | 0.011-0.026  (5) |
|  | RSA | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) |
|  | SAB | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) |
|  | SAF | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) | (0) |
|  | SAH | 2.7-16.6  (13) | 8.4-68.4  (13) | 0.038-0.183  (7) | 0.153-0.701  (16) | 1.21-6.92  (16) | 0.088-0.737  (16) | 4.18-118  (16) | 14.8-396  (16) | 0.104-2.16  (16) |

**Table S7.** Dry deposition fluxes (µmol m-2 d-1) of NO3-, NH4+, SP and soluble (s-) and total (t-) TMs to the North and South regions of the ETNA estimated using our baseline calculation (Base; Table 6), uncertainties estimated based on numbers of observations (num) and uncertainties based on the inter-quartile range of observed concentrations (IQ).

|  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Season | Region | Mode | NH4+ | NO3- | SP | s-Fe | s-Al | s-Mn | t-Fe | t-Al | t-Mn |
|  |  |  |  |  |  |  |  |  |  |  |  |
| MAM | N | Base | 1.1 | 15.1 | 0.014 | 0.034 | 0.24 | 0.016 | 1.5 | 5.1 | 0.03 |
|  |  | num | 0.8-1.3 | 12.5-17.8 | 0.010-0.018 | 0.029-0.040 | 0.20-0.28 | 0.013-0.018 | 1.1-1.9 | 3.7-6.6 | 0.02-0.03 |
|  |  | IQ | 0.7-1.9 | 10.0-20.8 | 0.009-0.018 | 0.022-0.052 | 0.17-0.46 | 0.008-0.022 | 0.7-2.8 | 1.6-9.5 | 0.01-0.05 |
|  | S | Base | 0.8 | 8.4 | 0.011 | 0.016 | 0.12 | 0.012 | 1.0 | 3.8 | 0.02 |
|  |  | num | 0.5-1.2 | 6.0-10.8 | 0.006-0.016 | 0.010-0.021 | 0.07-0.16 | 0.008-0.016 | 0.7-1.4 | 2.5-5.0 | 0.01-0.03 |
|  |  | IQ | 0.6-1.4 | 6.9-10.9 | 0.007-0.014 | 0.010-0.021 | 0.07-0.17 | 0.007-0.020 | 0.6-1.6 | 2.0-5.7 | 0.01-0.03 |
|  |  |  |  |  |  |  |  |  |  |  |  |
| JJA | N | Base | 2.6 | 13.7 | 0.021 | 0.048 | 0.37 | 0.023 | 3.5 | 13.2 | 0.06 |
|  |  | num | 2.2-3.0 | 11.6-15.8 | 0.018-0.025 | 0.041-0.056 | 0.31-0.43 | 0.019-0.026 | 2.9-4.2 | 10.8-15.5 | 0.05-0.07 |
|  |  | IQ | 1.5-4.1 | 12.0-20.0 | 0.016-0.030 | 0.032-0.069 | 0.20-0.56 | 0.011-0.037 | 2.2-7.6 | 8.4-27.1 | 0.04-0.14 |
|  | S | Base | 0.6 | 5.0 | 0.006 | 0.007 | 0.06 | 0.005 | 0.8 | 3.0 | 0.02 |
|  |  | num | 0.2-1.1 | 1.0-9.0 | 0.001-0.012 | 0.004-0.010 | 0.04-0.08 | 0.003-0.008 | 0.3-1.3 | 1.0-4.9 | 0.01-0.03 |
|  |  | IQ | 0.3-1.2 | 3.6-7.8 | 0.004-0.011 | 0.006-0.010 | 0.04-0.10 | 0.004-0.015 | 0.7-1.9 | 2.2-6.7 | 0.01-0.04 |
|  |  |  |  |  |  |  |  |  |  |  |  |
| SON | N | Base | 0.8 | 10.1 | 0.011 | 0.028 | 0.22 | 0.025 | 2.2 | 7.4 | 0.03 |
|  |  | num | 0.6-0.9 | 8.2-11.9 | 0.010-0.013 | 0.024-0.033 | 0.19-0.26 | 0.021-0.029 | 1.9-2.6 | 6.2-8.5 | 0.03-0.04 |
|  |  | IQ | 0.4-1.5 | 8.0-17.3 | 0.008-0.018 | 0.017-0.066 | 0.10-0.51 | 0.013-0.050 | 0.9-5.5 | 2.7-23.7 | 0.02-0.11 |
|  | S | Base | 0.8 | 5.4 | 0.009 | 0.010 | 0.09 | 0.009 | 1.0 | 3.5 | 0.02 |
|  |  | num | 0.6-1.0 | 4.3-6.5 | 0.007-0.011 | 0.008-0.011 | 0.07-0.10 | 0.008-0.011 | 0.8-1.1 | 2.9-4.0 | 0.02-0.02 |
|  |  | IQ | 0.4-1.4 | 3.3-8.7 | 0.006-0.014 | 0.006-0.019 | 0.05-0.18 | 0.006-0.020 | 0.6-1.9 | 2.3-7.4 | 0.01-0.04 |
|  |  |  |  |  |  |  |  |  |  |  |  |
| DJF | N | Base | 1.4 | 7.3 | 0.016 | 0.078 | 0.77 | 0.065 | 9.3 | 31.1 | 0.16 |
|  |  | num | 1.1-1.7 | 5.8-8.8 | 0.013-0.020 | 0.066-0.090 | 0.66-0.89 | 0.056-0.075 | 7.9-10.7 | 26.4-35.8 | 0.13-0.18 |
|  |  | IQ | 0.6-2.1 | 4.3-12.3 | 0.011-0.026 | 0.023-0.133 | 0.13-1.34 | 0.013-0.145 | 2.6-21.6 | 8.4-76.5 | 0.04-0.38 |
|  | S | Base | 1.0 | 9.0 | 0.024 | 0.045 | 0.36 | 0.048 | 4.7 | 19.3 | 0.09 |
|  |  | num | 0.6-1.5 | 6.3-11.7 | 0.015-0.033 | 0.037-0.053 | 0.30-0.43 | 0.040-0.056 | 3.9-5.5 | 16.2-22.3 | 0.08-0.10 |
|  |  | IQ | 0.6-1.8 | 6.4-11.5 | 0.019-0.031 | 0.029-0.069 | 0.23-0.64 | 0.039-0.071 | 3.7-8.5 | 15.6-33.8 | 0.07-0.17 |

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