Supplementary information for:

Methods for identifying aged ship plumes and estimating contribution to aerosol exposure downwind of shipping lanes

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Instrument set-up and meteorological condition

Fig. S1 shows a sketch of the complete measurement setup and the flow configuration used in the Falsterbo measurement campaigns. Data from several of the instruments in Fig. S1 will be presented in a companion article (S. Ausmeel et al., *Ship plumes in the Baltic Sea Sulphur Emission Control Area: Chemical characterization and contribution to coastal aerosol concentrations*, manuscript in preparation, 2019b).

The chemical composition of sampled particles was evaluated with a Soot Particle Aerosol Mass Spectrometer (SP-AMS, Aerodyne Research Inc.). (Onasch et al., 2012) In addition to the AMS measurements, black carbon (BC) content was measured with optical absorption methods, using a seven wavelength Aethalometer (model AE33, Magee Scientific) (Drinovec et al., 2015) and a 637 nm Multi Angle Absorption Photometer (MAAP, Thermo Fisher Scientific) (Müller et al., 2011), both with a sample time of one minute. A potential aerosol mass oxidation flow reactor (PAM OFR) (Kang et al., 2007;Lambe et al., 2011) was alternately connected before the AMS, SMPS, and Aethalometer to simulate atmospheric aging. For the gaseous aerosol compounds, CO2 concentration was measured with a non-dispersive infrared gas analyser (LI-COR LI840) and SO2 was measured using a UV fluorescent monitor (Environnement S.A AF22M). 1.1 Subsection (as Heading 2)

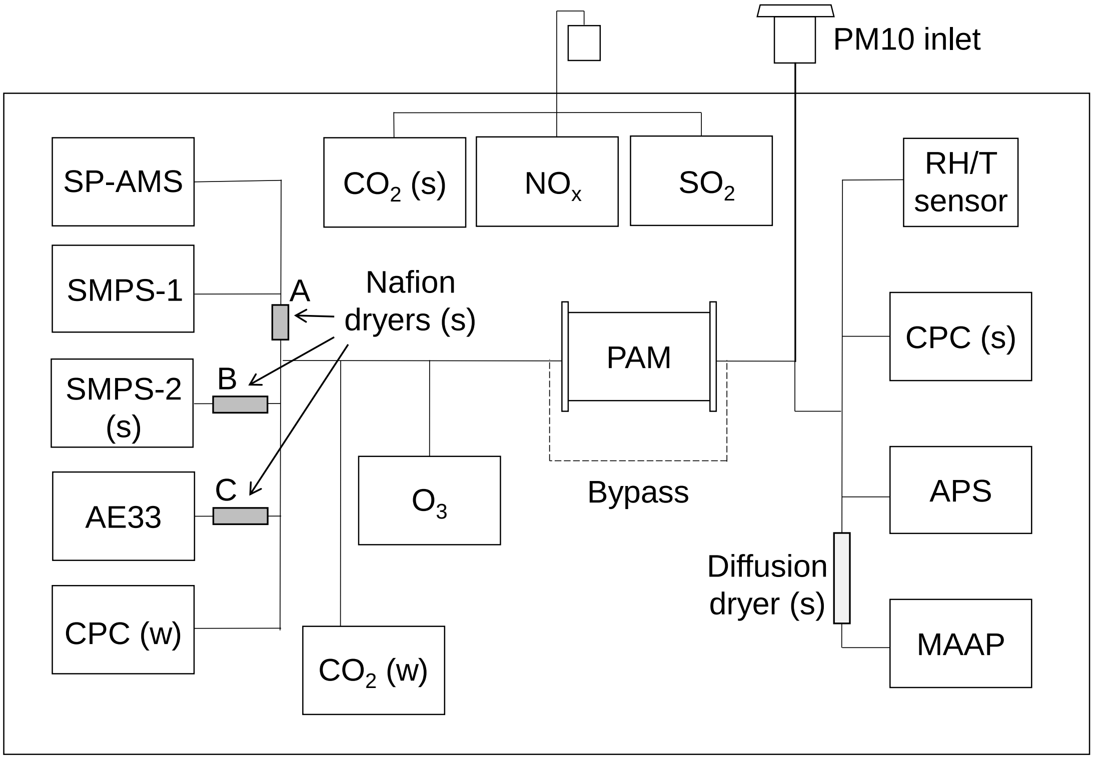

Figure S1. Measurement setup. The symbol (s) indicates configuration used only during the summer and (w) only during the winter. Dashed line shows the bypass flow excluding the PAM oxidation flow reactor from the sampling line. For the membrane (Nafion) dryers, the letters correspond to the size dependent particle penetration for each drier shown in Fig. S2.

CO_2 concentration enhancements due to ship plumes were below the detection limit of the monitor used, which means that emission factors likely cannot be calculated for ship plumes 7-20 km downwind of the shipping lane. In summary, for the MAAP (detection limit, DL, of < 50 ng m^{-3}), APS (DL 0.001 cm^{-3}), CO_2 (DL < 1 ppm), and SO_2 (DL < 1 ppb) monitors, the concentrations from ship emissions were at all times undistinguishable from the background levels. These data sets were not analyzed further.

Meteorological conditions, both averages and extremes, are presented in Table S1.

Table S1. Meteorological conditions during measurement campaigns, average, lowest and highest values.

Parameter	Winter (18/1 - 3/3)			Summer (16/5 – 7/7)		
	Min	Av ± stdv	Max	Min	Av ± stdv	Max
Temperature (deg. C)	-5.2	2.5 ± 2.4	6.6	9.2	17.0 ± 2.9	25.9
RH (%)	63	87.8 ± 7.9	99.0	38	77.7 ± 11.8	99.0
Wind speed (m/s)	0.3	7.5 ± 3.4	17.0	1.0	6.4 ± 2.8	15.0
Sunlight (h/day) [a]	0	1.93 ± 2.44	9.37	0	7.96 ± 4.57	15.6
Precipitation (mm/day)	0	0.8 ± 1.4	5.9	0	1.6 ± 3.8	31.8

[a] Direct sunlight, i.e. not cloudy.

Particle penetration of membrane dryers

Table S2 presents the specifications for each dryer used in the summer campaign to dry the aerosol particles before sampling with some of the particle instruments. Letters A-C correspond to the dryers shown in the illustration of the Falsterbo measurement setup in Fig. S1. The flows for which the losses are characterised (Table S2) were the same flows as used in the field measurements. The aerosol used for the characterization was polydisperse ammonium sulphate in lab room air. The resulting losses, as a fraction of the total particle concentration, are shown as function of particle size in Figure. S2.

Table S2. Specifications of dryers used in Falsterbo, letters A-C correspond to the driers in Fig. S1.

Dryer	Serial no.	Dryer length (Wiedensohler et al.)	Flow used (lpm)
A	MD-110-12E-S (082913-02-18)	30.5	1.1
B	MD-110-24S-4 (1060301)	61	0.3
C	MD-110-24S-4 (0860108)	61	3

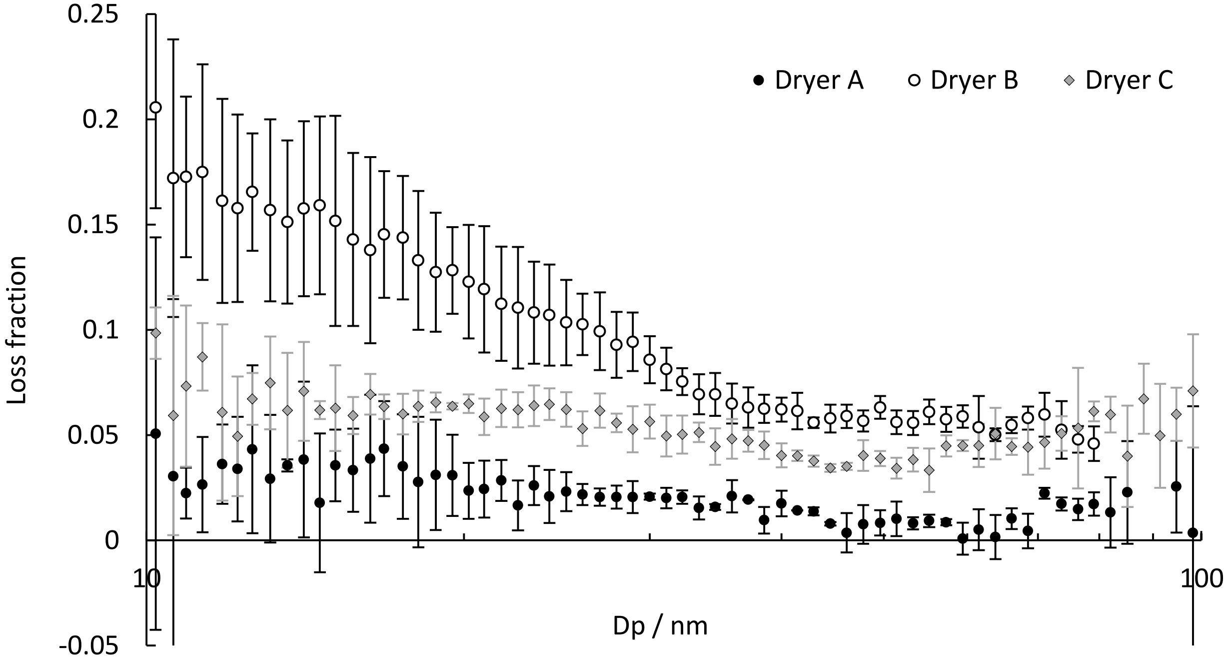

Figure S*2*. Fraction of total particle concentration lost due to diffusion in three dryers, as a function of particle diameter, D_p. Error bars indicate one standard deviation from 2-3 measurements.

Log-normal fit to ship particle size distributions

Four or five modes are used in the log-normal fit (Table S3) of the average size distribution plume since it seems that the typical size distribution contains a smaller and a larger sized nucleation mode (mode no. 1 and 2, < 30 nm diameter), and a smaller and larger sized Aitken mode (30 to 100 nm diameter). A majority of the ships do not produce the lower sized nucleation mode, why the median size distribution does not contain this first mode. The other modes are often all present at the same time and the larger particles could arise due to coagulation in an aerosol with a high concentration of smaller particles or due to emissions of relatively large primary soot particles. The uncertainties for the size distribution are large for the particles in the upper Aitken mode (80 to 100 nm diameter) and the accumulation mode (>100 nm diameter) due to low numbers counted in the SMPS and also due to large variation between individual ships.

Table S3. Lognormal fit parameters for the average and median size distribution of the detected ship emission particles, during winter (n=61) and summer (n = 8) respectively.

Parameter	Winter		Summer	
	Median size distr.	Average size distr.	Median size distr.	Average size distr.
$N_1 \pm \Delta N$ (cm^{-3})	-	603.66 ± 615.34	1302.72 ± 1054.28	916.82 ± 731.82
$N_2 \pm \Delta N$ (cm^{-3})	584.75 ± 290.75	890.06 ± 351.06	942.05 ± 278.05	775.03 ± 329.97
$N_3 \pm \Delta N$ (cm^{-3})	222.33 ± 167.67	214.66 ± 171.66	293.57 ± 84.57	603.19 ± 188.81
$N_4 \pm \Delta N$ (cm^{-3})	7.79 ± 11.21	35.46 ± 32.60	17.49 ± 23.51	13.58 ± 25.42
$N_5 \pm \Delta N$ (cm^{-3})	-	-	9.68 ± 7.62	22.74 ± 21.56
GMD_1 (nm)	-	9.95	10.74	14.13
GMD_2 (nm)	19.04	21.46	29.84	27.49
GMD_3 (nm)	34.17	41.52	51.82	46.65
GMD_4 (nm)	86.29	98.04	84.16	84.31
GMD_5 (nm)	-	-	125.73	112.16
σ_1	-	1.30	1.35	1.35
σ_2	1.51	1.60	1.35	1.35
σ_3	1.39	1.45	1.22	1.35
σ_4	1.37	1.32	1.10	1.10
σ_5	-	-	1.13	1.27

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