Figure S1: Hourly mean Hg$^0_{air}$ concentrations at (a) Mace Head, (b) Cabo Verde Observatory, (c) Cape Point and (d) Amsterdam Island. The mean ± 1 standard deviation as well as the range of the hourly observations are shown in each panel. Note that the extent of both the x- and y-axis differs across the four panels.

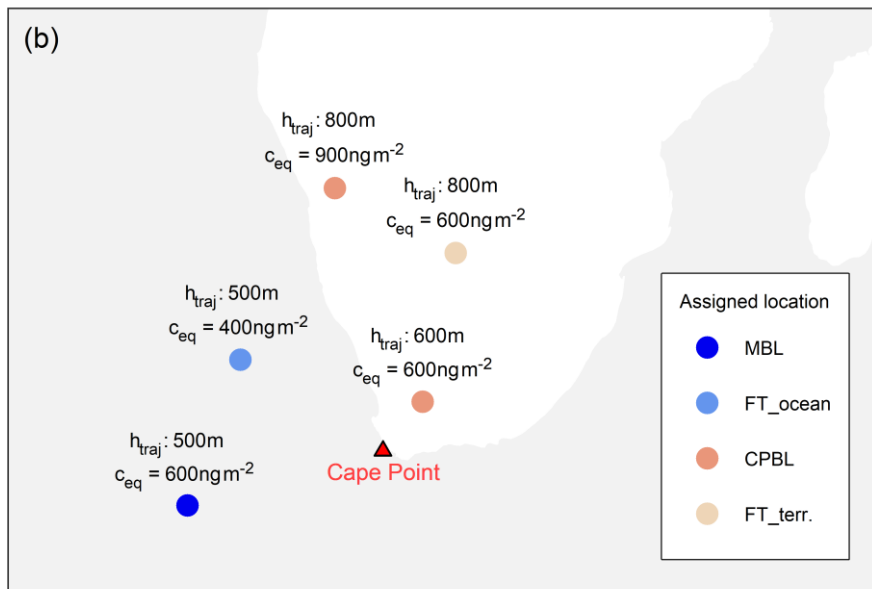
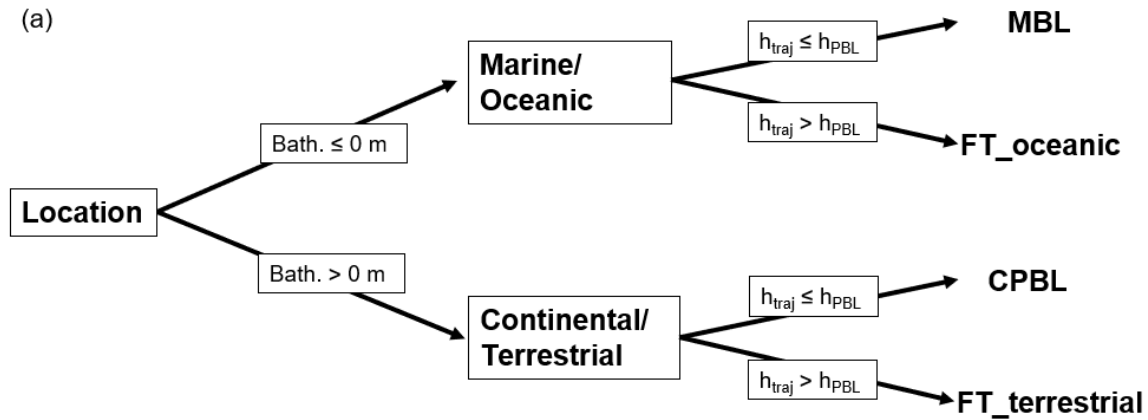



Figure S2: Illustration of the trajectory-segment categorisation applied in the study. (a) Framework of the trajectory-segment categorisation applied in the study. Bathymetry denotes the GEBCO-derived bathymetry at the location of the trajectory, "h_{traj}" the height above ground/sea level of the trajectory segment, and "h_{PBL}" the ERA5-obtained PBL height at the instance (i.e., the time and geographical location) of the trajectory segment. Segments over the ocean at a height equal to or below the PBL height are assigned MBL, while those over the ocean above the PBL are FT_ocean. Similarly, segments over land at or below the height of the PBL are assigned CPBL, while those over land above the PBL are FT_terrestrial. (b) Randomly generated trajectory points to illustrate the categorisation

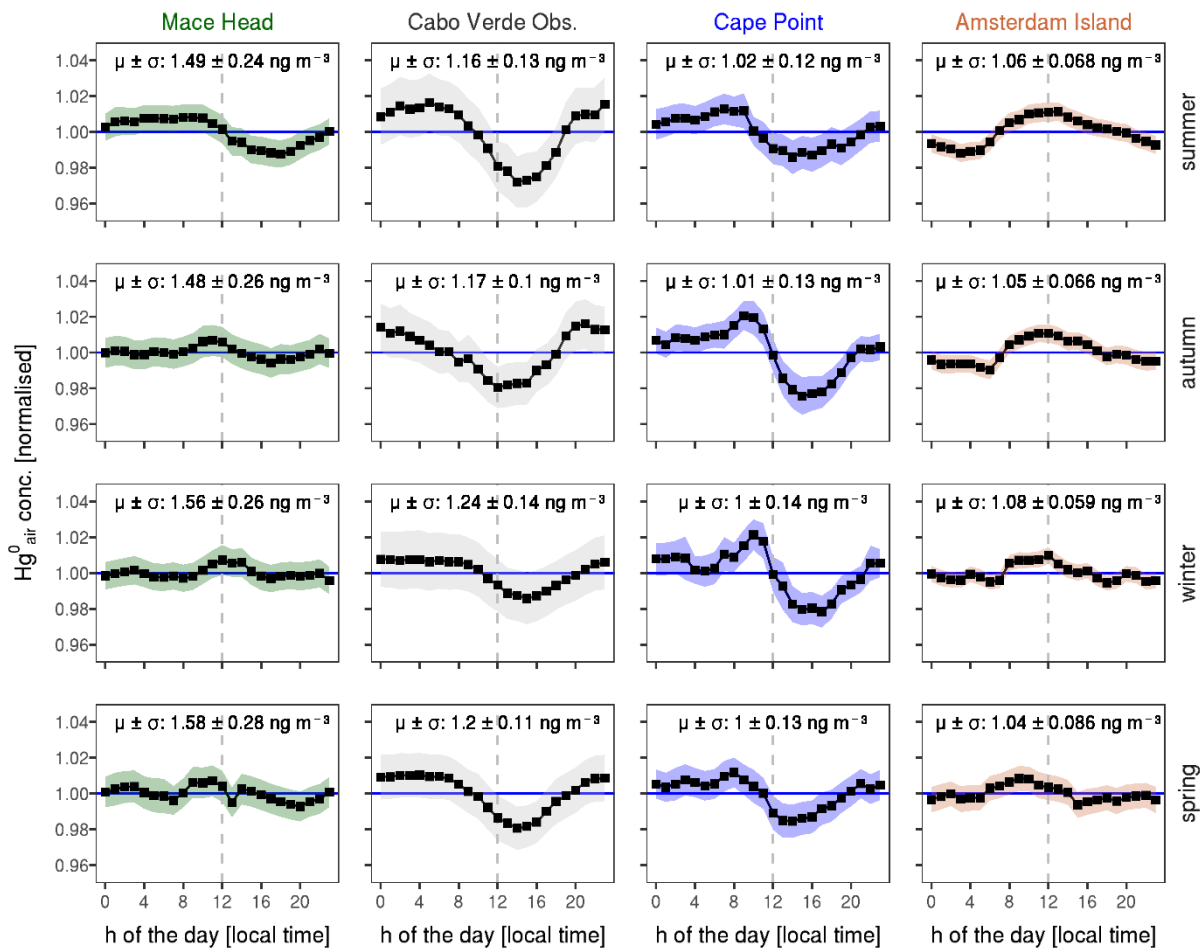



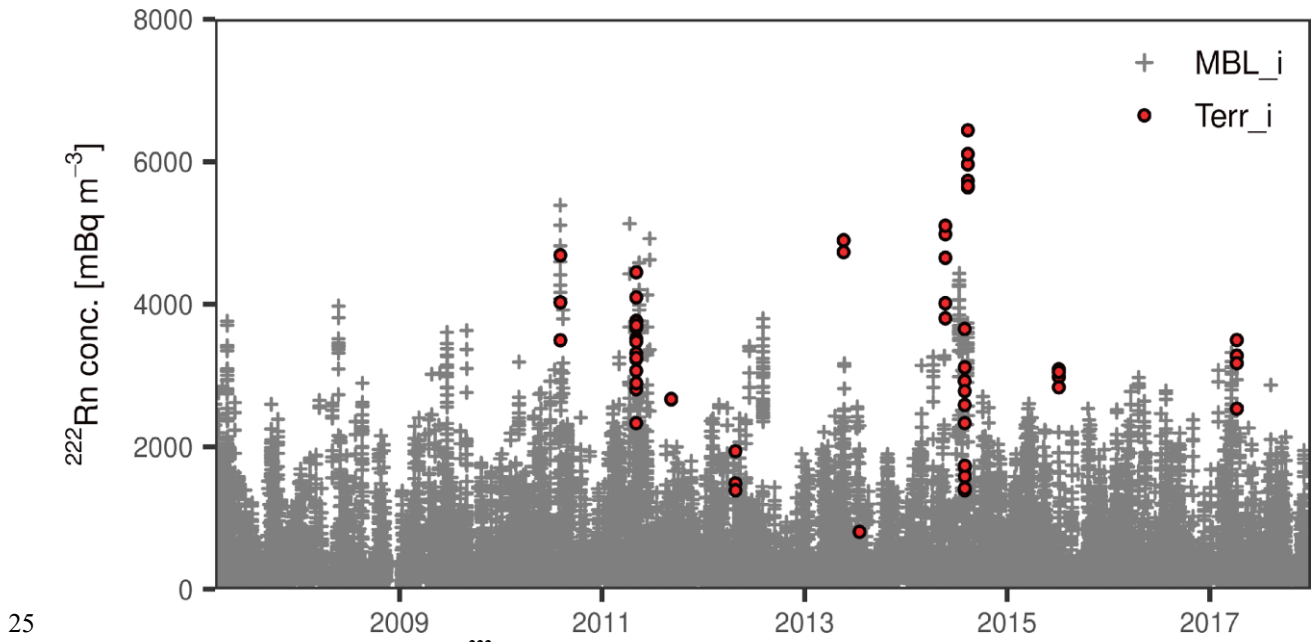
Figure S3: Normalised mean atmospheric Hg⁰ diurnal variation at the fur study sites in different seasons. Summer, autumn, winter and spring refer to Jun–Aug, Sep–Nov, Dec–Feb and Mar–May, respectively, for Mace Head and Cabo Verde Observatory, and to Dec–Feb, Mar–May, Jun–Aug and Sep–Nov, respectively, for Cape Point and Amsterdam Island. The mean ± the standard deviation of the hourly observations in each season is shown in the panel. The shaded area shows the mean ± 2 times the standard error. The horizontal line represents the normalised mean value (i.e., 1 ng m⁻³) and the dashed vertical line is plotted along 12:00 local time.

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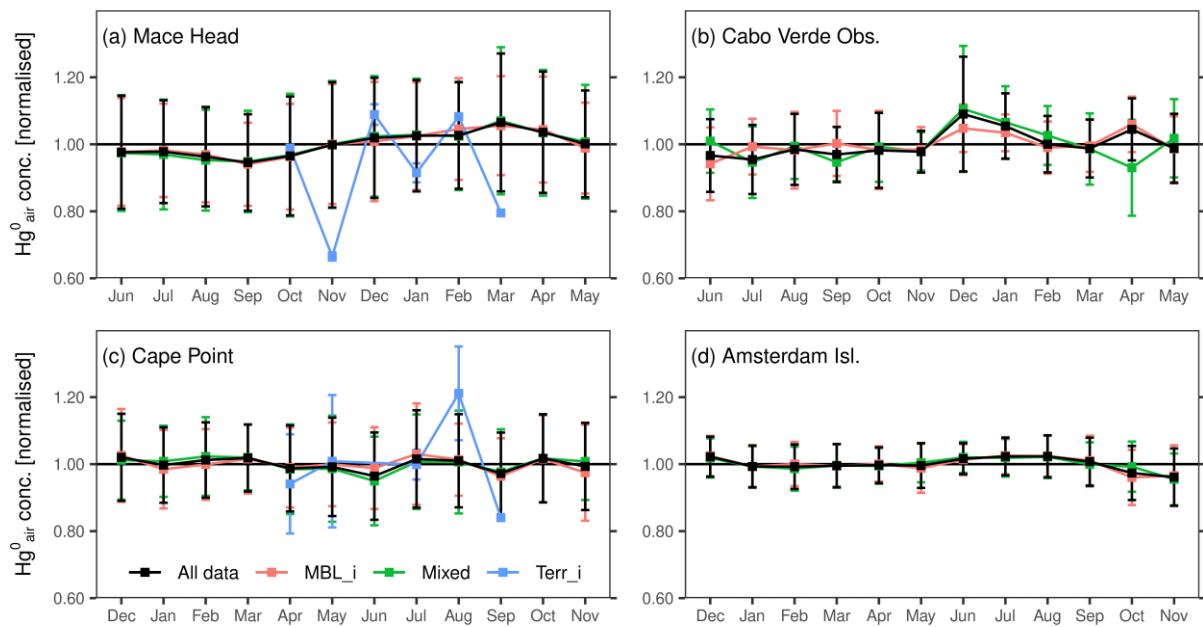
Figure S4: Time series of hourly mean ^{222}Rn concentrations at Cape Point, with the data points distinguished between MBL_i and Terr_i.

30 **Figure S5: Similar to Fig. 2 in the main text but also including the MBL_i, Terr_i and Mixed Hg$^0_{air}$ datasets for each site. The mean, standard deviation and proportion of hourly observations assigned to each category is provided in Table S1 in this Supplement.**

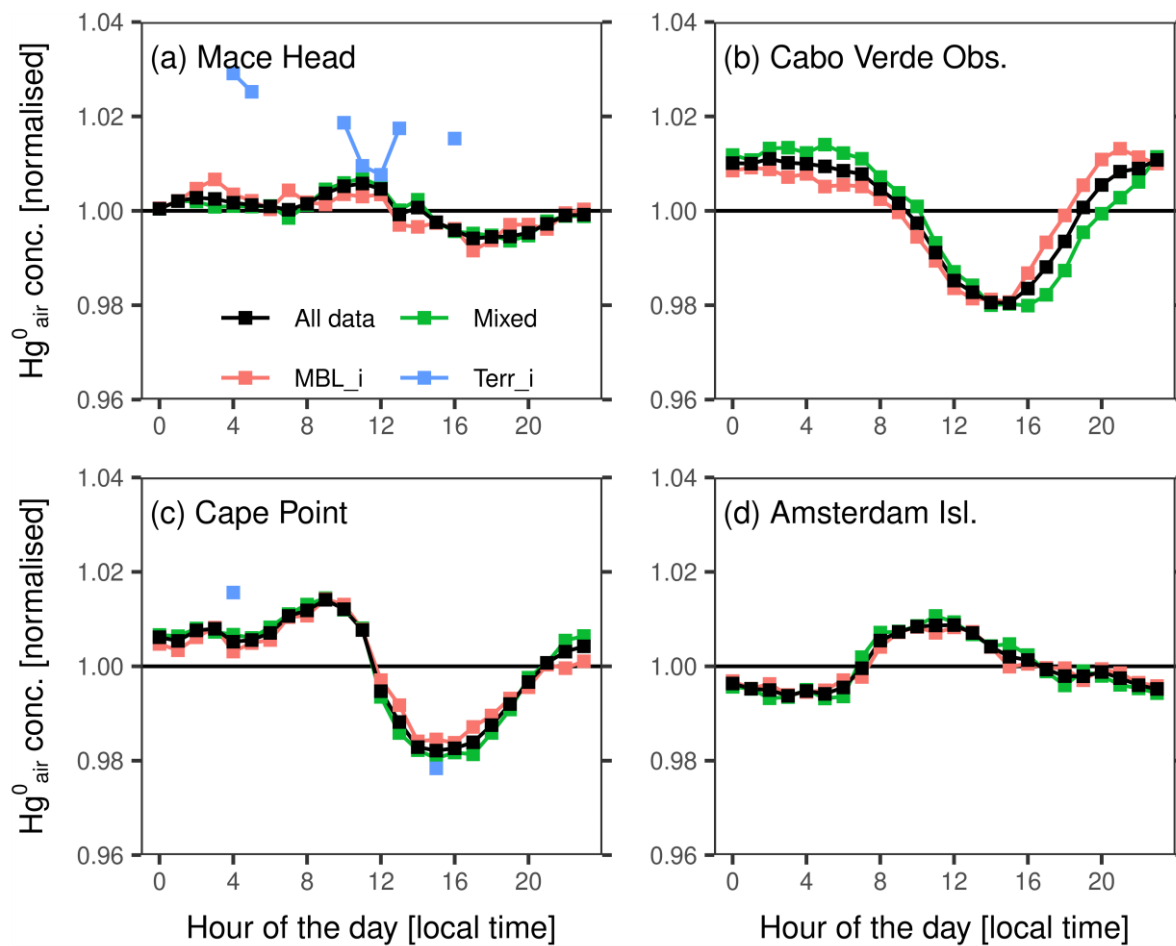



Figure S6: Similar to Fig. 3 in the main text but also including the MBL_i, Terr_i and Mixed Hg^0 datasets for each site. The mean, standard deviation and proportion of hourly observations assigned each category is provided in Table S1 in this Supplement.

Table S1: Hg^0_{air} mean concentration, standard deviation as well as proportion of the data from the full the full data ("All data") classified into the group, for the groups MBL_i, Terr_i and Mixed, at the four study sites.

Group	Mean [ng m^{-3}]	Std. dev. [ng m^{-3}]	Proportion of All data [%]
		Mace Head	
All data	1.53	0.26	
MBL_i	1.53	0.24	28.73
Mixed	1.52	0.27	71.24
Terri_i	1.70	0.35	0.03
		Cabo Verd Obs.	
All data	1.19	0.13	
MBL_i	1.19	0.11	53.39
Mixed	1.21	0.14	46.61
Terri_i	–	–	0
		Cape Point	
All data	1.01	0.13	
MBL_i	1.01	0.12	42.95
Mixed	1.00	0.13	56.96
Terri_i	1.07	0.20	0.09
		Amsterdam Isl.	
All data	1.06	0.07	
MBL_i	1.06	0.07	55.74
Mixed	1.06	0.07	44.26
Terri_i	–	–	0

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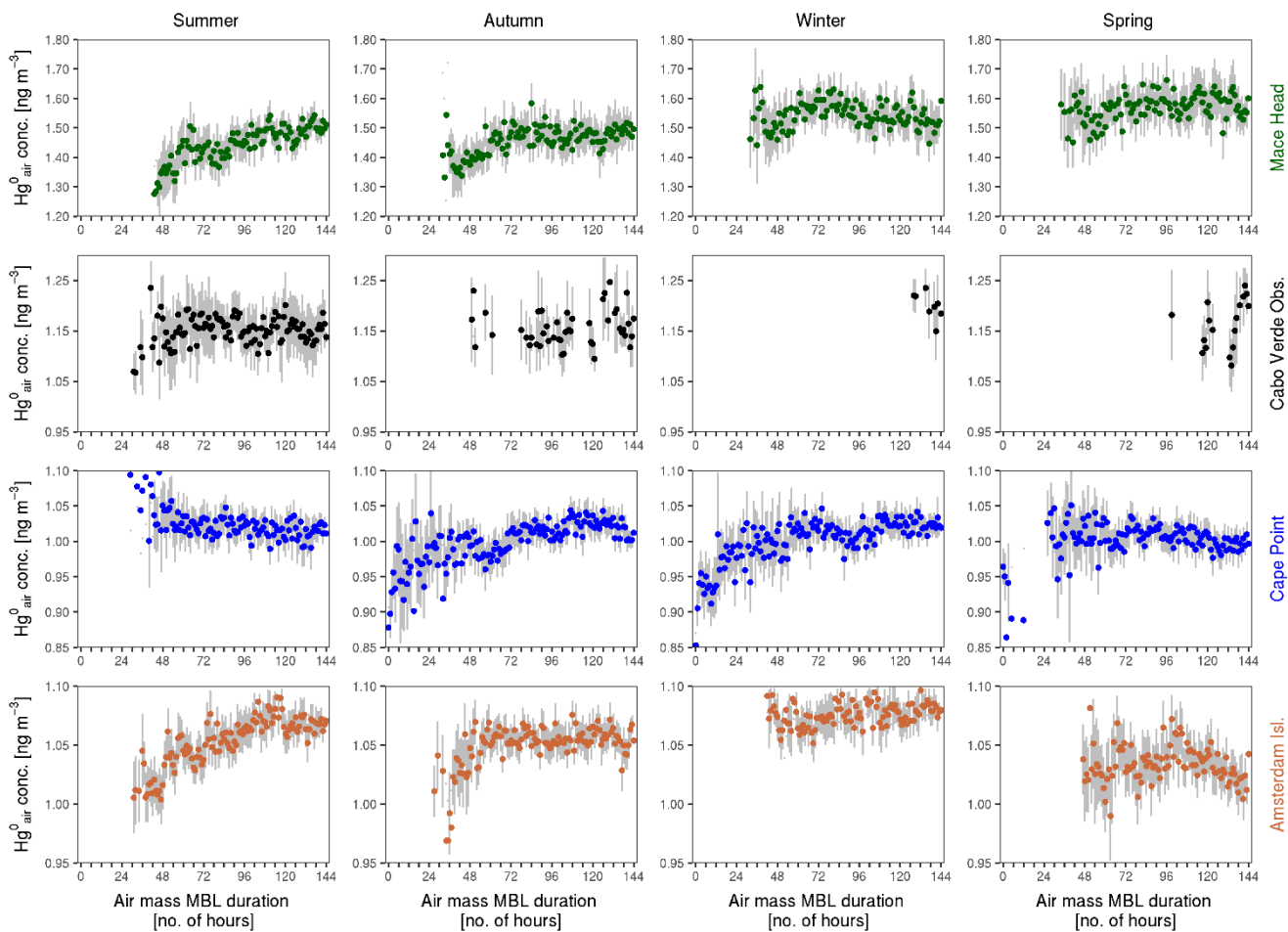
Figure S7: Seasonal variation in the relationship between mean Hg$^0_{air}$ concentration and air mass recent MBL residence time at Mace Head, Cabo Verde Observatory, Cape Point and Amsterdam Island. Summer, autumn, winter and spring refer to Jun−Aug, Sep−Nov, Dec−Feb and Mar−May, respectively, for Mace Head and Cabo Verde Observatory, and to Dec−Feb, Mar−May, Jun−Aug and Sep−Nov, respectively, for Cape Point and Amsterdam Island. The bars show 2 times the standard error of the means. The extent of the y-axis differs across the four sites.