Methods / Trajectory analysis

Lagrangian trajectory models are widely used to in investigate source-receptor relationships of atmospheric constituents based on air-mass movement from gridded meteorological data (Fleming et al., 2012). The method involves the calculation of an air parcels movement from the receptor site (i.e. back in time) which yields the back-trajectory of the parcel (Draxler and Hess, 1998). Individual trajectories are then matched with observed atmospheric constituents as measured at the receptor site to derive a spatial pattern on the air-mass origin in relation to the respective concentration of the constituents at the receptor site.

In this work, the air-mass back-trajectory analysis was conducted using the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) version 4.9 (Draxler and Hess, 1998; Stein et al., 2015). The HYSPLIT model was run using the ensemble method 7 days back in time. The ensemble method will offset the meteorological grid by one grid point in the horizontal and 1% of the surface pressure in the vertical. This will produce 27 back-trajectories for all possible offsets in the horizontal and vertical thus accounting for uncertainties in the gridded meteorological data. The meteorological data used for the trajectories was the NCEP/GDAS dataset with a 1° horizontal resolution and 23 pressure levels (Kanamitsu, 1989).

The back-trajectory analysis was done by assigning the aerosol properties as measured at the arrival time and assigning those properties to the grid-cell centres that that the trajectory-path traversed. This allows for a footprint to be constructed using the measured aerosol properties at the different receptor sites as

$$C_i = \left(\frac{\sum_{n=1}^{N} c_n w_n}{\sum_{n=1}^{N} w_n}\right)_i \qquad (1)$$

In Eq. (1), C_i is the footprint of the aerosol property c_n weighted by w_n comprising N trajectory passes through the grid location i. The grid comprised 40962 geodesic cells for better statistics near the pole. Due to multiple receptor locations, the weights used were inverse distance travelled from the receptor point. Thus, the receptor location closest to a grid cell will weigh the grid cell the most which is justified since the trajectory path becomes more uncertain with greater distance travelled (Draxler and Hess, 1998). The aerosol properties c_n investigated were light absorption coefficients (σ_{AP}), light scattering coefficients (σ_{SP}) and single scattering albedo (ω_0).

In the Arctic, and in general, few sources of atmospheric pollutants should be present high above ground. Therefore, C_i was restricted to back-trajectory altitudes below 500 meters above ground level. An altitude of 500 metres was chosen as a compromise since there are not be enough trajectories within the planetary boundary layer (PBL) for reasonable statistics, especially given the low PBL depth in the Arctic.

Results / Trajectory analysis

Figure X shows the footprint of light absorption coefficient, light scattering coefficients and single-scattering albedo. The figure was constructed using weighted arithmetic means using Eq. (1). Weighing was done using the reciprocal trajectory distance from the receptor point. Thus, the station closest on the map will influence the footprint the most.

Figure (Xa) depicts the σ_{AP} footprint of the six Arctic stations. The figure shows that during the measurement period considered, air-masses that originate from the east (i.e. from Russia) were associated with higher σ_{AP} than those associated with air masses originating over North-America. Model results imply that the Russian Arctic is a substantial pathway for poleward transport of BC into the Arctic whereas North America is not (Huang et al., 2010). This suggests that the general

trend of Fig. (Xa) holds true. In this study, the lowest σ_{AP} could be found over an area covering southern Greenland expanding out over the ocean to the Svalbard archipelago (i.e. the Norwegian Sea).

The method used for the footprint does not include any deposition, just airmass location history. Therefore, a source region that is far away from any of the measurement stations is likely to be affected by removal processes, such as wet deposition, along the way. It remains unclear if removal processes, or the method used here, is the reason that Europe does not show up in Fig. (Xa) although the area should be a substantial contributor to light absorbing species in the Arctic ((Huang et al., 2010; Koch and Hansen, 2005) . Again, uncertainties in the method increase with distance to any of the measurements stations. However, the area east of the Caspian Sea with higher values of σ_{AP} both showed up in the individual footprints of the stations Pallas, Alert and to some extent Tiksi.

Figure Xb shows the Arctic footprint of σ_{SP}. Again, the area associated with low σ_{SP} values is southern Greenland, and to some extent the Norwegian Sea. Similar to the footprint of σ_{AP}, σ_{SP} is also higher over the Eastern Arctic that comprise northern Russia. Furthermore, a similar area of elevated values of σ_{SP} can be found between Scandinavia and the Caspian Sea. Another area of somewhat elevated levels of σ_{SP} is western Canada.

In Fig. (Xc), the Arctic footprint of ω_0 is depicted. The figure implies that the highest values of ω_0 in the high Arctic are to be found over the Arctic ocean with darker aerosol over the continents. The mean ω_0 over the Arctic ocean seems to be quite constant; roughly 0.95. However, eastern Canada seems to be the area with the highest ω_0 values whereas northern Greenland in an area associated with low ω_0. The low ω_0 of northern Greenland is not likely to be true since ω_0 values close to 0.8 are more often observed at populated continental sites than in the high Arctic.

Figure X: Footprint of **(a)** light absorption coefficients (σ_{AP}), **(b)** light scattering coefficients (σ_{SP}) and **(c)** single-scattering albedo (ω_0) using data from the six Arctic stations at a wavelength of 700 nm. The figure comprise back-trajectories where the trajectory altitude was less than 500 metres above ground level. Red stars mark the locations of the respective stations. Latitude and longitude grid lines are spaced 15º and 30º apart, respectively. Grid cells that have fewer than 5 trajectory overpasses have been omitted.

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