

Assessing the relative contributions of transport efficiency and scavenging to seasonal variability in Arctic aerosol

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ABSTRACT

Regional aerosol concentrations are governed by an evolving balance between aerosol sources and sinks. Here, a simple technique is described for making estimates of the extent to which seasonal aerosol variability is controlled by wet scavenging rather than the efficiency of transport from pollution source regions. Carbon monoxide (CO) is employed as an assumed passive tracer of pollution transport efficiency, to which the magnitude of aerosol light scattering is compared. Because aerosols, unlike CO, are affected by wet scavenging as well as transport efficiency, the ratio of short-term perturbations in these two quantities provides a measure of the relative roles of these two processes. This technique is applied to surface measurements in the Arctic at Barrow, Alaska (71°N) for the decade between 2000 and 2009. What is found is that a well-known seasonal cycle in 'Arctic Haze' is dominated by variability in wet scavenging. Crossing the freezing threshold for warm rain production appears particularly critical for efficiently cleaning the air.

1. Introduction

In 1906, George C. Simpson remarked that 'All who have travelled in Arctic regions know the peculiar haze which fills the air when the temperature falls very low and gives the 'cold' aspect to Arctic scenes. Such a haze, which is not a mist or fog, was frequent during the winter in Karasjok [69N in Norway]. On the other hand, at the end of the summer the air reached a degree of transparency which I have never seen equalled in any other place'. (Simpson, 1906; Garrett and Verzella, 2007)

'Arctic Haze' is now known to originate primarily from lower-latitude industrial activities (Rahn et al., 1977; Shaw, 1982; McConnell et al., 2007). Aerosol concentrations in the lower few kilometres of the Arctic atmosphere build between approximately December and April and rapidly clean out in summer (Shaw, 1995; Polissar et al., 1999; Quinn et al., 2007). The topic has renewed focus due to concerns that aerosols may be playing a role in rapid Arctic climate change (Menon et al., 2008). Suggested mechanisms include perturbations by aerosol of solar reflection and thermal radiative emission (Garrett and Zhao, 2006; Lubin and Vogelmann, 2006), the direct radiative impacts of haze aerosol (Shaw et al., 1993), and contributions to surface

melting from soot deposited on snow (Warren and Wiscombe, 1980; Hansen and Nazarenko, 2004). It is thought that carefully chosen policies aimed at aerosol control may be applied to mitigation of Arctic warming and sea-ice melting (Shindell, 2007; Quinn et al., 2008).

If aerosols are indeed having a significant radiative impact, the aerosol seasonality that Simpson and others noticed must be placed within the context of a pronounced annual solar cycle. The Arctic is dark in winter, and obviously this influences the relative contributions of long-wave and short-wave radiation to aerosol-induced perturbations (Garrett et al., 2002; Law and Stohl, 2007). It will be the mapping of the aerosol and solar cycles that determines the nature and timing of the ultimate radiative perturbation.

Of course, the solar cycle is easily explained, but the annual aerosol cycle is not. Aerosol variability is determined by a balance between evolving sources and sinks. In the case of the Arctic, there are two processes that are normally highlighted: The air is unusually polluted in winter because air is transported more efficiently to polar regions from industrialized regions; the Arctic Front is further south and pollution is trapped vertically by a low-level inversion (Heintzenberg and Larssen, 1983; Raatz and Shaw, 1984; Stohl, 2006). On the sink side, the winter atmosphere is particularly stable and the temperature cold, so precipitation rates are low and this diminishes wet scavenging of pollution (Shaw, 1995).

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What remains to be quantified is the relative impacts of transport efficiency and scavenging to the observed aerosol cycle (Quinn et al., 2007). Does Arctic air near the surface clean out in summer primarily because the Arctic Front has receded and export is more efficient, or rather due to accelerated deposition to the surface by increased snow and rain? The answer is not obvious. In principle, the relative roles of each component process could be assessed with a detailed transport model (Crutzen and Lawrence, 2000; Rasch et al., 2000; Shindell et al., 2008). However, it is much less clear how to observationally validate the extent to which model parametrizations of sources and sinks are correctly represented. This paper introduces a simple method for separating the respective roles of transport efficiency and scavenging in surface data, applied specifically to measurements obtained at Barrow, Alaska.

2. Mechanisms

Aerosol concentrations in a pollution plume can become lowered through some combination of mixing with surrounding air, ‘wet scavenging’ by precipitation, or direct deposition to the surface through ‘dry scavenging’. Following an air parcel in a Lagrangian sense, instantaneous variability in aerosol concentration χ follows

$$\frac{d \ln \chi}{dt} = -\psi, \quad (1)$$

where $\psi = \psi_m + \psi_w + \psi_d$ is the instantaneous removal rate through combined mixing, wet and dry deposition, respectively. (Note that ψ_m can be negative depending on local concentration gradients.) If, instead, a given instant and place is considered, directly calculating χ requires consideration of mixing and removal over all past trajectories that contribute to the air parcel in a given location.

Mixing processes can be addressed using tracer transport models based on meteorological re-analyses. These show that interactions of near-surface Arctic air with polluted lower latitudes are about twice as slow in summer as in winter (Stohl, 2006). Effectively, summertime exchange is less ‘efficient’.

Dry deposition of aerosol is a very slow removal process in the Arctic due to a relative absence of vegetation. Larger heavy metal aerosol may be removed through dry deposition over an atmospheric lifetime $1/\psi_d$ of order 1 week (Vinogradova, 2000), but haze aerosol are removed more slowly because particles in the characteristic 0.1 to 1 μm diameter size range tend to slide past rather than directly impact surfaces (Slinn, 1977). We assume here that dry scavenging has a negligible contribution to the observed seasonal cycle in aerosol concentrations.

Provided precipitation is present, wet scavenging is the most efficient aerosol removal mechanism. Aerosol nucleate cloud droplets, which are then carried to the surface through collisions and subsequent coalescence with falling raindrops, or through

the riming of falling snowflakes. The removal rate is

$$\psi_r = \alpha P/L, \quad (2)$$

where P and L are the column-averaged precipitation rate and amount of condensate, respectively, and α is the solubility of aerosol in cloud (Crutzen and Lawrence, 2000; Garrett et al., 2006). Assuming a monthly averaged precipitation rate of 1 mm d^{-1} based on observations at Barrow, Alaska (Zhao and Garrett, 2008), and a liquid water path of order 100 g m^{-2} ($L = 0.1 \text{ mm}$) (Curry et al., 1996), a typical rain-out time for perfectly soluble aerosol in the Arctic summer is on the order of 1 hr. A similar removal rate might apply even during very cold conditions, since precipitation rates and water paths approximately scale (Garrett et al., 2002; Zhao and Garrett, 2008). However, this rate would be an upper limit. If there is snowfall without aggregation or riming, aerosol lifetimes can be closer to 1 month (Davidson et al., 1987; Vinogradova, 2000). The reason is that, in cold conditions, precipitation particles can be solid and grow by vapour diffusion rather than collisions. In this case, a precipitation particle may contain only a single aerosol when it hits the ground, rather than the millions normally associated with a raindrop.

3. Methods

Rather than explicitly calculate removal rates along parcel trajectories, here we quantify scavenging using a more indirect approach (Garrett et al., 2006). In essence, combustion produces both aerosols and gases. If concentrations of an aerosol species can be compared to concentrations of an insoluble and chemically inert gas, what is provided is an indicator of the extent to which an air mass has been cleaned by wet scavenging. If the ratio is low, polluted air has been cleaned through scavenging rather than mixing.

For example, we previously applied this approach to co-located MODIS retrievals of cloud properties and transport model output of CO as an inert pollution tracer (Avey et al., 2007). In this case, elevated tracer model CO concentrations immediately downwind of Northeastern North America were associated with smaller retrieved cloud droplet effective radii, presumably due to an aerosol indirect effect on cloud properties. However, 3 d downwind of the continent the association became very weak. The explanation provided was that, through wet scavenging, rain had cleaned the atmosphere of aerosol pollution while the carbon monoxide (CO) was left behind.

In Garrett et al. (2006), wet scavenging was assessed using airborne measurements of aerosol between 0.1 and 1 μm diameter. Measured concentrations of CO, adjusted for some background value, were adopted as the insoluble, inert gas. As a by-product of combustion, CO near sources tends to strongly correlate with concentrations of combustion aerosol with sizes between 0.1 and 1 μm (Longley et al., 2005). Aerosol in this size range can be assumed to nucleate droplets in clouds (Dusek et al., 2006).

Further, CO is almost completely insoluble (Crutzen and Lawrence, 2000) and it is long-lived compared to the time scales that were considered. Garrett et al. (2006) found that when aerosol concentrations were low compared to CO, nearby clouds were heavily precipitating.

In the Arctic-based study described here, the measurable aerosol species to which CO concentrations are compared is the light scattering coefficient σ_{sp} (units Mm^{-1}) of dry aerosol smaller than $1 \mu\text{m}$ diameter. Light scattering correlates well with Arctic cloud properties (Garrett et al., 2004), probably because scattering is most efficient in the $0.1\text{--}1.0 \mu\text{m}$ size range (Seinfeld and Pandis, 1998) where Arctic haze aerosol tend to be concentrated (Leaith et al., 1989) and cloud nucleating aerosol are found (Dusek et al., 2006).

An important added consideration for the Arctic is that, while aerosol and CO have different atmospheric lifetimes, both can be longer lived than the time it takes for air to transit the Arctic. Pollution levels in the Arctic are always some combination of 'first pass' short-term pollution events and a time-dependent 'background' representative of older pollution recycled from extra-Arctic latitudes. This background value must be determined and subtracted to obtain the perturbation associated with recent pollution transport events (e.g. $\Delta\text{CO} = \text{CO} - \text{CO}_{bkgd}$). Then, in the absence of aerosol removal mechanisms other than transport, CO and aerosol perturbations should track each other. In principle, ΔCO can be used as an indicator of the efficiency of dynamic motions at transporting atmospheric aerosol perturbations $\Delta\sigma_{sp}$ from recent pollution sources.

So, while ΔCO and $\Delta\sigma_{sp}$ are both affected by transport, $\Delta\sigma_{sp}$ is also influenced by the potential rapid removal of aerosol to the surface through wet scavenging. Based on these considerations, the relative values of the two quantities follow

$$S = \frac{\Delta\sigma_{sp}(\text{location})}{\Delta\text{CO}} = f \frac{\Delta\sigma_{sp}(\text{source})}{\Delta\text{CO}}, \quad (3)$$

where the scavenging ratio S is an indicator of the fractional extent f to which the source relationship between $\Delta\sigma_{sp}$ and ΔCO has been altered by aerosol scavenging. If measured values of S are low, this implies heavy scavenging.

The described method for assessment of scavenging is simple but approximate. It is assumed that the relationship between $\Delta\sigma_{sp}$ and ΔCO at source regions is a constant. While there is certainly variability between individual combustion sources, there is atmospheric smoothing through mixing during transport to the Arctic, such that measured anthropogenic pollution represents an aggregate of many sources. That said, regional pollution sources do in fact shift some depending on the season (Polissar et al., 1999). The effect on $\Delta\sigma_{sp}/\Delta\text{CO}$ variability in the Arctic is unknown but assumed to be small.

Also, there are potential contributions from biomass burning, which can be associated with particularly high Arctic aerosol and CO levels (Novelli et al., 2003; Warneke, 2010). In general, the impact of biomass burning on surface measurements is small.

Although forest fires themselves peak in summer, injection tends to be into the upper atmosphere and poleward transport is along upward sloping isentropes (Stohl, 2006). Perhaps surprisingly, forest fire chemical tracers measured at the surface at Alert and Barrow do not peak in summer, but rather in winter and spring when the aerosol chemical signature is dominated by anthropogenic pollutants (Quinn et al., 2007). Of course, mixing may well introduce biomass aerosol into the boundary layer on occasion. In this study, we do not exclude strong biomass events from the analysed data set; biomass aerosol are also subject to wet scavenging. However, to avoid introducing potential bias from a small number of extreme biomass events, we focus on the median rather than the mean of each statistical cohort examined. Thus, the climatologies presented here are more indicative of a typical rather than an average scenario.

We also assume that processes other than mixing and scavenging are sufficiently slow as to be unimportant. This is in fact an approximation. First, aerosol grow in cloud droplets through aqueous phase chemical reactions. However, growth will be fastest where gas-phase concentrations are highest nearer lower-latitude sources (Schwartz and Freiberg, 1981). While scavenging and mixing can occur at any point along the transport pathway, we assume aqueous phase reactions are effectively complete close to sources, and have approached some equilibrium well before measurements are obtained in the Arctic.

More importantly, we assume that CO associated with short-term pollution events is not significantly oxidized in the time it takes for it to traverse the distance between source regions and the Arctic. CO is oxidized through reaction with the hydroxyl radical OH. OH is produced by the photolysis of ozone and drives a CO seasonal cycle with a winter minimum and a summer maximum. At the surface in northern latitudes, Duncan et al. (2007) show that, in January, CO lifetimes are in excess of 100 d. In July, lifetimes are as short as weeks in Western Europe and the United States, but are at least 40 d northward of 50° and greater than 100 d over Canada and Northern Europe, perhaps because cloud cover is nearly ubiquitous in the Arctic in summer. By comparison, residence times in the Arctic of air near the surface are approximately 1 week in winter and 2 weeks in summer (Stohl, 2006). By mass continuity, the implication is that oxidation lifetimes are normally much longer than the time it takes for CO to be transported to the Arctic. In our analysis here, we assume that oxidation controls the observed seasonality in Arctic background concentrations of CO, and only transport controls short-term perturbations. That said, in summer, residence (and, by continuity, transport) times are long and oxidation times relatively short. Taking 2 months as the oxidation time and 2 weeks as the transport time, approximately one quarter of a short-term CO perturbation might become lost to photolysis during summer-time poleward transit. In spring and fall, we estimate loss is perhaps 5%. As we will show, even though assuming zero loss is not entirely justified, it does not change this article's primary conclusion.

In effect, it is implicitly assumed here that the aforementioned assumptions combine to have a signature that is weak compared to the scavenging signature we intend to isolate. The advantage of making these approximations is that it is only clear-air measurements at a single location that are required to estimate the scavenging contribution, and no *a priori* knowledge of clouds, precipitation or mixing along the transport pathways. It is by using proxy variables and making approximations that the scavenging problem becomes tractable.

4. Measurements

The data used for this study is from the National Oceanic and Atmospheric Administration Global Monitoring Division Barrow (NOAA GMD BRW) site near Barrow, Alaska (71.32°N, 156.62°W). We use a decade of data spanning the years between 2000 and 2009.

Meteorological state variables measured at GMD BRW are described by Herbert et al. (1986). Measurements of aerosol scattering σ_{sp} are from the green (550 nm) channel of a TSI Inc. 3563 3-wavelength nephelometer (Anderson and Ogren, 1998). Upstream of the nephelometer, aerosols were dried then passed through one of two (1 and 10 μm) impactors. To omit events linked to long-range desert dust transport, only measurements made with the 1 μm impactor are considered here. Events possibly associated with local pollution from the City of Barrow are also removed.

Quasi-continuous measurements of CO were made using gas chromatography with hot mercuric reduction detection (Trace Analytical, Inc., model RGA3). Detector non-linear response was corrected using a piecewise linear interpolation between

three CO-in-air mixtures (Scott-Marrin, Inc.) ranging from ~ 60 to 200 ppb. Four to five mixing ratios were determined per hour. An expert system evaluated the chromatographic parameters and rejected data with parameters not within the accepted range. All other results were retained and summed into hourly and monthly averages. Hourly averaged in situ CO (from 1992 to 2000) agree to 1.4 ± 3.2 ppb with grab samples collected within the hour and measured at NOAA/ESRL in Boulder, Colorado.

Our goal here is to obtain values for a short-term perturbation in total CO concentrations and σ_{sp} that is distinguishable from a background concentration, where the background is constant over timescales longer than the perturbations of interest. Here, a quantified background level is based on short-term minima in BRW data. The minimum is defined within consecutive 2-week intervals as the lower 5% quantile. If less than 60% of hourly data points within the interval satisfy the aforementioned data quality flags for σ_{sp} and CO, then no background or perturbation values were calculated for the interval.

As shown in Fig. 1, background values exhibit strong seasonal cycles. In fact, in winter and spring, slowly varying background σ_{sp} and CO signals are often of greater magnitude than the shorter-term perturbations that we isolate: the Arctic doesn't regularly 'clean-out'—what background that exists is often polluted itself.

Figure 2 shows seasonal changes at Barrow for monthly mean values of the local precipitation rate P between 2000 and 2003 (from Zhao and Garrett, 2008), monthly median values of the short-term perturbations in CO and aerosol light scattering σ_{sp} between 2000 and 2009, and the ratio $\Delta\sigma_{sp}/\Delta\text{CO}$.

Median values in monthly light scattering perturbations $\Delta\sigma_{sp}$ follow an opposing cycle to local precipitation rates, with a

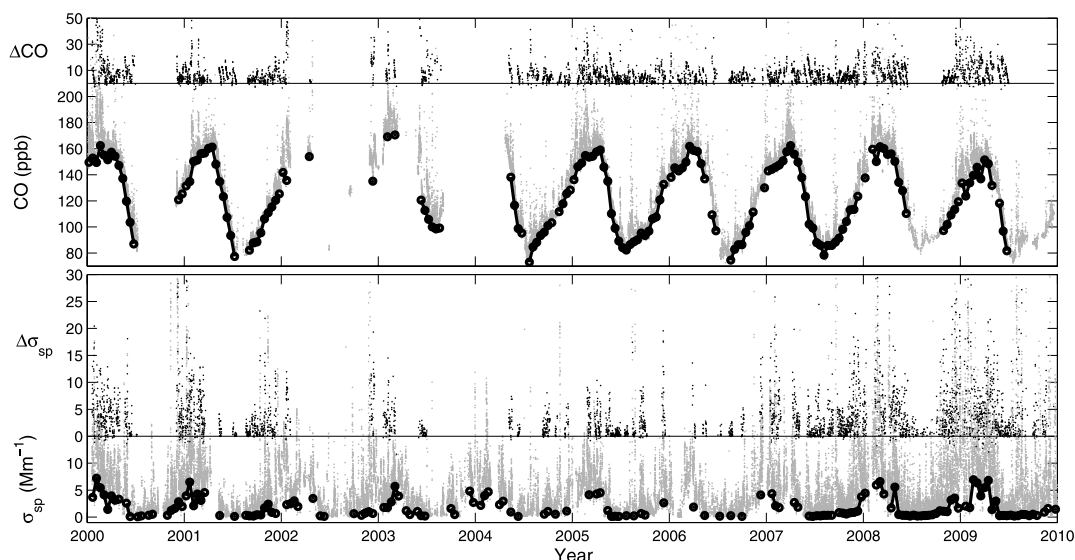


Fig. 1. Monthly σ_{sp} and CO concentrations at BRW obtained between 2000 and 2009 (grey dots). Black lines with circles represent estimated background values within consecutive 2-week intervals, representative of slowly varying trends. Black dots represent the short-term perturbation with the background subtracted.

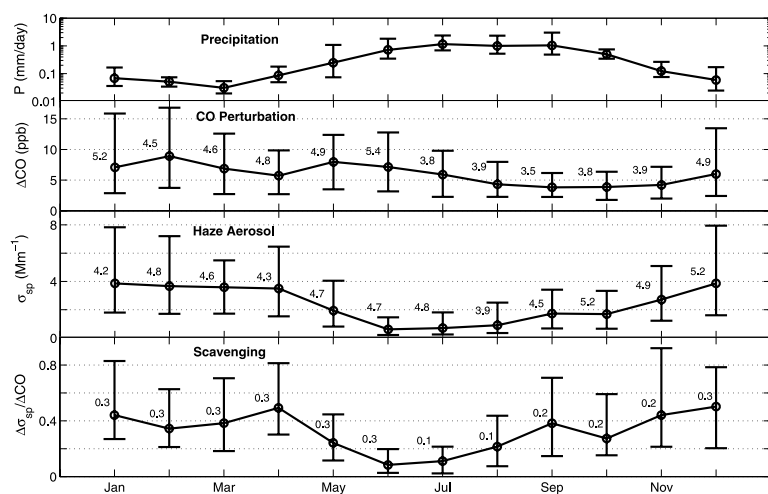


Fig. 2. For data obtained at Point Barrow, monthly changes in precipitation rate between 2000 and 2003 (Zhao and Garrett, 2008), and for the years 2000–2009, monthly median values for hourly perturbations in CO, aerosol light scattering, and the monthly median of the scavenging ratio where relative hourly perturbations in ΔCO and $\Delta\sigma_{sp}$ are coincident and $\text{CO} > 3.2$ ppb. Ranges specify upper and lower quartiles. Numbers represent the number of hourly averaged data points, in thousands.

pronounced minimum between June and October that is as much as 10 times less than concentrations in winter. The cycle in median values of ΔCO is much smaller. The apparent efficiency of pollution transport weakens gradually, by about a factor of two, between winter and late summer.

For calculations of median values of hourly $S = \Delta\sigma_{sp}/\Delta\text{CO}$ for each month, only coincident hourly averaged events with $\Delta\text{CO} > 3.2$ ppb are considered in order to obtain sufficiently high signal to noise. Results are only weakly sensitive to this threshold choice. The calculated scavenging ratio is up to four times lower in mid-summer than in winter and early spring. The concurrent mid-summer reduction in ΔCO is comparatively small implying that, in mid-summer at least, CO transport efficiency to Barrow is not strongly inhibited. The implication then is that wet scavenging is what controls the very low haziness characterizing June and July. Even if transport efficiency were underestimated because we ignored oxidation of CO en route to the Arctic, then the importance of wet scavenging in the aerosol cycle would be even higher.

In September and October, however, the role of transport and scavenging is reversed. The scavenging ratio S has values that are similar to those seen in winter and early spring, while values

of ΔCO are at a minimum. It appears that transport efficiency is more important than scavenging at maintaining intermediately clean conditions during the fall.

It is revealing here to compare values of S with local meteorological conditions for a low scavenging season between November and April, and a high scavenging season in June and July (Fig. 3). During the low scavenging months, the most typical values of S are approximately $0.4 \text{ Mm}^{-1} \text{ ppb}^{-1}$, independent of local temperature or relative humidity. In June and July, however, values of S are approximately $0.1 \text{ Mm}^{-1} \text{ ppb}^{-1}$, when concurrent values of relative humidity are near liquid saturation and temperatures are above freezing.

These results are interesting because, for one they suggest that there are distinct most-typical values of S that characterize both the low and high scavenging seasons. Absent scavenging, a value for $\Delta\sigma_{sp}/\Delta\text{CO}$ of $0.4 \text{ Mm}^{-1} \text{ ppb}^{-1}$ looks broadly representative of mid-latitude pollution sources. Also, the data suggest that what is critical for a high scavenging mode to dominate is that high relative humidity air becomes exposed to temperatures above freezing. As discussed earlier, precipitation may be present, but wet scavenging will only be high if precipitating hydrometeors grow through collision mechanisms rather than

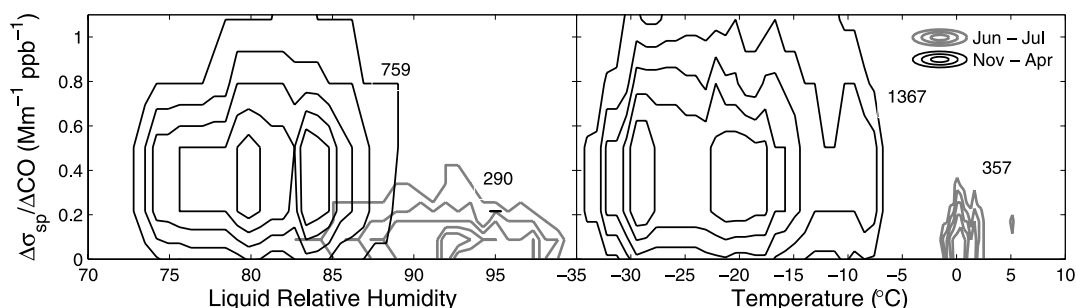


Fig. 3. For the years 2000–2009 at BRW, the probability distribution for hourly averaged surface measurements of scavenging $\Delta\sigma_{sp}/\Delta\text{CO}$, and the meteorological variables of the relative humidity with respect to liquid and air temperature. Black contours represent a nominally polluted season between November and April, and grey contours a scavenged season in June and July. Numbers represent the number of hourly averaged data points.

vapour deposition. There are two possibilities here. Riming involves a combination of vapour deposition and droplet collisions. More efficient though is warm rain and drizzle production, which is due exclusively to droplet collision-coalescence and requires liquid clouds with temperatures higher than 0 °C.

5. Conclusions

It has long been noted that air in the Arctic is hazy in winter and early spring and pristine in summer. The results presented here suggest that, at least at Barrow, Alaska, and averaged over a decade long period between 2000 and 2009, transport efficiency and scavenging play unequal roles in controlling this seasonal cycle. In the fall, the air is relatively clean primarily because pollution transport to Barrow is less efficient. But during mid-summer, the air is exceptionally transparent, and it is wet scavenging of aerosol that appears to account for the absence of Arctic Haze.

We were able to arrive at this conclusion using a very simple technique for approximating the extent to which aerosol concentrations have been influenced by scavenging. The method requires no measurements of clouds or precipitation. Nor does it require detailed assumptions of the scavenging processes involved. Instead, the relative influence of scavenging and transport is implicit in short-term perturbations from more slowly varying background concentrations of an aerosol quantity and a pollution transport tracer. Here, we used light scattering coefficient perturbations for the aerosol quantity and CO perturbations as the transport tracer. These quantities are normally included in long-term measurement campaigns at other remote locations (Novelli et al., 1992; Delene and Ogren, 2002). Thus, the technique may have broad applicability.

One surprising result from our study is that efficient wet scavenging is associated with warm, moist air with temperatures near to or above the freezing point. In general, air is cleaned more efficiently by rain than snow. In the Arctic, the freezing point appears to serve as a ‘scavenging point’ that is only passed in the mid-summer months. This suggests that it may be important for models of the role of aerosol in future Arctic climate change to consider not only future changes in pollution sources, but also the future evolution of anthropogenic aerosol sinks. If rain is required for efficient wet scavenging, then plausibly a shift to a warmer moister Arctic regime will lead to both clearer Arctic air and greater surface aerosol deposition.

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