

Seasonal Changes in the Transport of Pollutants into the Arctic Troposphere- Model Study

A. Klonecki,¹ P. Hess,

National Center for Atmospheric Research, Boulder, CO, U.S.A.

D. Blake

University of California, Berkeley, CA, U.S.A.

¹) Now at Laboratoire des Sciences du Climat et de l'Environnement, Gif-sur-Yvette, France

Abstract

In association with the Tropospheric Ozone Production about the Spring Equinox (TOPSE) measurement campaign a regional episodic chemical transport model (CTM) is used to study the mechanism of transport of pollutants from their primary Northern Hemisphere emissions regions into the remainder of the troposphere. The model, whose domain includes the high- and mid-latitudes of the Northern Hemisphere, was run from December 1999 till July 2000 with 56 chemical species. The model simulates the strong, seasonal cycle in the transport of mid-latitude pollution into the high latitudes, in agreement with early studies on Arctic haze. The simulated seasonal cycle for CO and hydrocarbons agrees well with measurements from the TOPSE aircraft measurement campaign conducted in the Canadian Arctic during the winter and spring of 2000. In this study we analyze the transport mechanisms that are responsible for this strong seasonality and identify and quantify the impact of various emission regions throughout the troposphere. Episodic transport events over the deep Arctic are examined during the time period of the TOPSE campaign.

A series of simulations with passive and short lived diagnostic tracers labeled by the region and potential temperature of the emissions allows a quantification of the effect of diabatic heating in simulated transport pathways. Examining the transport from an isentropic perspective explains many of the transport differences found between different seasons and different emission regions. The diagnostic tracers also allow the contribution of different emission regions to tracer concentrations in the mid- and high latitude troposphere to be quantified. In the high latitudes during winter, in agreement with previous studies, the European and Siberian sources are the largest contributors to pollution in the lower troposphere due to favorable large-scale transport patterns, the low potential temperatures at the emission source, and the presence of a cold, stable boundary layer that facilitates diabatic cooling en route to the Arctic. The majority of North American sources are located further south than the European emissions and are on average emitted at higher potential temperatures. Due to their predominant transport over the Atlantic, the North American emissions experience strong diabatic heating due to the general instability in the air column over the relatively warm Atlantic Ocean and the large precipitation events connected with frontal passages off the coast of North America. As a result the North American emission sources reach the Arctic significantly more diluted than the European sources and at higher altitudes. During the summer months the pollution from all emission regions is more likely to be diabatically transported to higher potential temperatures and diluted. In addition, the direct transport of pollutants into the Arctic is less frequent due to the differences in the large scale circulation patterns.

1 Introduction

A number of measurement studies conducted in remote locations north of the polar front have indicated a dramatic seasonal variation in the concentration of pollutants originating from midlatitude industrial centers. Hydrocarbons (HC) concentrations reach a maximum during the winter months and decrease throughout the spring with a minimum during summer and fall. Measurements taken during the TOPSE (Tropospheric Ozone Production about the Spring Equinox) campaign [*Atlas et al.*, this issue] over N. America between 40N and 85N confirm this trend and show that the buildup of HC maximizes at the high latitudes, well away from large emission sources (see figure 1).

It has been suggested that with the return of sunlight in the spring this buildup of HC could lead to a "photochemical bloom" and subsequent rapid ozone production. One of the major objectives of the Tropospheric Ozone Production about the Spring Equinox (TOPSE) campaign was to document the winter-spring transition in tropospheric photochemistry and to investigate the contribution of the possible bloom in chemistry to the observed seasonal ozone maximum. The TOPSE campaign consisted of a series of aircraft flights from Denver, CO to Thule Greenland (76N, 68W) or Alert Canada (82.5N, 62W) between February and late May [*Atlas et al.*, this issue].

The seasonal increase in the concentration of pollutants has been especially well documented for the high Arctic, where pronounced pollution events are frequently present during winter and spring. These events, which have come to be known as Arctic Haze, manifest themselves in sharp, episodic decreases in the visibility over the lower troposphere. They are caused by large concentrations of aerosols thought to primarily have an anthropogenic origin. Due to its dramatic nature, Arctic Haze has been studied since the 1970's through a number of measurement programs [e.g., *Barrie and Hoff*, 1985, *Barrie et al.*, 1994]. Less well documented is the seasonal buildup of hydrocarbons in the Arctic (e.g., in Alaska: *Blake and Rowland*, [1986], on Spitsbergen: *Solberg et al.*, [1996]). The seasonality in hydrocarbons has been documented to extend considerably south of the Arctic, however. A number of studies (in the Canadian boreal forest: e.g., *Jobson et al.*, [1994] and *Young et al.*, [1997], over the North Atlantic from aircraft measurements [*Penkett et al.*, 1993], and over North America during TOPSE [*this issue*]) have shown that a strong seasonality in HC occurs south to the polar front, suggesting that HC go through a similar cycle as the Arctic Haze well south of the arctic region, with maximum concentrations in the winter or spring and minimum concentrations during summer. The amplitude of the cycle depends on the species and on the location. Note that the polar front usually lies considerably south of the arctic region, and separates mid-latitude airmasses from those of tropical and subtropical origin.

The buildup of HC north of the polar front and pollution events in the arctic region (connected with Arctic Haze) are often treated separately and yet similar processes are at least partially responsible for both phenomena. Both

are likely to be affected by the much slower winter removal rates: wet removal in the case of aerosols, and oxidation by OH or the hypothesized Br and Cl reactions in the case of HC. Moreover, during the winter months, large pollution sources are often located north of the polar front, while during the summer period these sources are isolated to the south. It has been also suggested in a number of studies [Barrie, 1986] that seasonal changes in the circulation of the lower troposphere can lead to large seasonal changes in the pollutant transport into the arctic region. For example, *Barrie* [1986] argues that the maximum transport to the Arctic takes place in winter, while in the summer this transport is much slower. *Lamarque and Hess* [this issue] also examine the build-up of pollutants during the Northern Hemisphere winter during TOPSE by showing how CO emitted from different emission regions builds-up and decays during the winter months.

In this study we use a three-dimensional chemical transport model to examine how seasonal changes in transport patterns affect the springtime transition in chemical concentrations, and more importantly, how and why the transport pathways from midlatitude pollution sources differ with season. We show that the region most affected by seasonal transport differences is the high latitude lower troposphere. While this region was not significantly sampled during the TOPSE campaign, the springtime transition in transport is also evident throughout the mid- and high-latitude troposphere.

In agreement with previous studies we find that the large-scale winter circulation is more conducive to strong poleward transport of Eurasian pollution than the circulation during the summer months. However we also find that large seasonal transport differences can also be attributed to changes in atmospheric heating and cooling between the winter and summer months. An analysis from the point of view of transport along surfaces of constant potential temperature (isentropic surfaces) provides a powerful mechanism to interpret the seasonal changes in the transport pathways into the high and mid-latitudes and their change with season. We also quantify the contribution of different emission regions to tracer concentrations throughout the mid and high latitude troposphere and interpret these contributions using the isentropic framework. The isentropic methodology has been previously applied to studies connected with the Arctic Haze [e.g., *Carlson*, 1981; *Iversen and Joranger*; 1985, *Raatz*, 1991], although on a more limited scale.

The paper is organized as follows. In section 2 we give the model description. In section 3 we compare the seasonal cycle of hydrocarbons in the model to the measurements, examine the large scale aspects of this seasonal cycle, and the evolution of episodic transport events as winter transitions into summer. The transport pathways of emissions are examined in section 4 using an isentropic framework and diagnostic tracers of anthropogenic emissions. We present our conclusions in section 5.

2 Model description

The HANK regional chemical transport model is described in *Hess et al.*, [2000] with recent changes outlined in the Appendix to this paper. Only a brief description of the model is repeated here. HANK is a chemical transport model (CTM) that belongs to a class of models often termed "off-line." These models do not calculate the dynamical fields themselves, but instead rely on meteorological models to provide fields necessary to describe the state of the atmosphere. HANK uses the fields calculated by the MM5 (PSU/NCAR Mesoscale Modeling System [*Grell et al.*, 1993a]) model. The advantage of using the MM5 rather than the standard meteorological analysis (e.g. NCEP [National Center Environmental Prediction) analysis) is that both models are run on the same horizontal and vertical grid (no interpolation is necessary), the output can be saved at higher spatial and temporal resolution than available using analyzed fields, and additional variables can be saved to recreate a consistent parameterization of subgrid processes. In addition the user has control over the choice of parameterizations that are used in the parent meteorological model.

In these simulations both the MM5 and HANK use a polar stereographic grid with the North Pole in the middle of the domain (the whole domain is shown for example in figure 3). Using this domain the model is able to simulate the transport of tracers by the midlatitude cyclones and transport from the primary emission regions which impact the concentrations of tropospheric tracers in the mid and high latitudes of the Northern Hemisphere. In addition the choice of the grid eliminates any numerical problems associated with transport across the pole (i.e., there are no singularities there). The horizontal resolution used here is fairly coarse, due to the long integration times, and is typical of a global CTM. The grid size at 60N is 243km. In order to be able to simulate the small scale vertical structure observed during other measurement campaigns [*e.g.*, *Ridley et al.*,] the vertical resolution was chosen to be fine- there are 39 levels between the surface and 100mb (the top of the model).

The HANK model uses the Smolarkiewicz advection scheme [*Smolarkiewicz* 1984], a different scheme from that used in the MM5. The MM5 advective scheme proves to be too diffusive for tracer transport. For both deep and shallow convective transport HANK uses the Grell convective parameterization scheme [*Grell*, 1993b], which is also employed in the MM5. For diffusive and boundary layer mixing the MRF counter gradient scheme was chosen for both the MM5 and HANK.

The MM5 model runs providing the input fields to HANK consisted of overlapping 30 hour segments. In each of these runs the initial six hours was discarded, allowing for model spin-up. The winds and other dynamical parameters calculated for the last 24 hours were then used to transport species in HANK. The NCEP aviation model provided the initial and boundary for these MM5 runs. In addition the winds in the MM5 were relaxed to the NCEP

analysis using the Newtonian relaxation approach [Grell *et al.*, 1993a]. The short integration times of the MM5 (30 hours) combined with the relaxation procedure ensure that the meteorological fields produced within the MM5 are close to the NCEP analysis.

3 Simulation and Measurements during winter and spring of 2000

The TOPSE campaign provided an extensive and unique data set of the composition of the winter and spring troposphere with measurements spanning the region from 40N (Boulder) to 82.5N (Alert). We begin by using this data set to evaluate the aspect of the model most relevant to this paper: the timing and magnitude of the simulated seasonal cycle for hydrocarbons and CO (section 3.1). We then proceed to analyze the seasonal change in the large scale distribution of CO and the meteorological factors that contribute to this change (section 3.2). Next we examine some of the episodic transport events simulated during the TOPSE period (section 3.3). In section 4 we quantify the differences in the seasonal transport using diagnostic tracers.

3.1 Seasonal Evolution of HC during TOPSE -Simulation and Measurements

To evaluate the model against measurements [Blake *et al.*, this issue], the simulated concentrations for various HC and CO are interpolated to the location and time of each measurement along the aircraft flight tracks. Figure 1 shows the median mixing ratios in the range of 1 to 6 km for ethene, ethane, propane and CO for four latitude bands for the seven deployments of the TOPSE campaign: 1) February 4-9, 2) February 21-27, 3) March 5-8, 4) March 19-26, 5) April 2-11, 6) April 23-30, 7) May 15-23. Note that initially the simulated concentrations tend to be somewhat high, while at the end of the campaign the modeled concentrations tend to be low. Nevertheless, the model generally reproduces the timing of the seasonal decrease, and even captures the relative maximum in many of the concentrations during the fourth deployment. In agreement with measurements the most rapid decrease in the concentration of the short lived ethene takes place early in the campaign. In both the model and measurements the largest decrease in the other species does not occur until later (after the fourth mission), with the seasonal decrease in the longer lived species, CO and ethane, being less pronounced. HANK also correctly captures the strong latitudinal gradient that exists during the first few deployments and its disappearance near the end of the campaign.

A comparison of the modeled and measured vertical profiles of propane and their evolution during the TOPSE campaign is depicted in figure 2. The last four deployments capture the large transition from winter to spring conditions. The model reproduces the seasonal change in concentration and vertical structure, capturing the transition

between the relatively high vertical propane gradient during deployment 4 to the very small vertical gradient present during deployment 7.

The agreement between the simulated and measured seasonality of these four species is generally good. The model accuracy is generally better than 25-30% (the emissions are probably not known to a higher accuracy), but in certain locations and times the model error is larger (e.g. CO is too low near the end of the campaign). Initially the error tends to be large due to an inadequate knowledge of the initial conditions. Nevertheless, qualitatively the meridional and vertical gradients, as well as the timing of the seasonal transitions for CO and the HC are reproduced correctly. This agreement gives us confidence that the model correctly captures the seasonal transition in transport to the arctic region.

3.2 The large scale seasonal evolution during TOPSE

The seasonal evolution of CO and HC along the TOPSE flight paths reflects the large scale changes in the simulated tracer concentrations which occur in the mid- and high latitudes during the springtime transition. We examine these changes in this section and place them in the context of large scale changes in the circulation. We focus on the lower troposphere with a particular emphasis on the arctic region. Previous studies and our model results show the lower Arctic is the region most affected by seasonal transport differences.

Figure 3 shows the seasonal evolution in CO on the 850mb pressure surface. The largest seasonal change at 850mb clearly occurs in the Arctic: the largest winter time concentrations occur there, while in June the lowest concentrations are simulated in the Arctic. Clearly the increase in CO oxidation with season is partially responsible for these changes. We show below, however, that changes in transport patterns also contribute strongly to this seasonal transition. Figure 3 offers a hint that the transport is important- transport of pollutants over the pole from Eurasia is clearly seen in the February and April CO distribution, but not in June. Strong transport from Eurasia to the Arctic during winter and early spring has been suggested in previous studies (e.g. Raatz and Shaw, 19984, Barrie 1986) and partially attributed to the changes in the location of the large-scale tropospheric pressure systems and the resulting circulation. For a qualitative picture, it is instructive to look at figure 4, which shows the model's monthly averaged geopotential heights at 850mb. During the winter months, the large strong low pressure systems over Iceland and the Aleutian islands and the two well developed continental high pressure systems over Siberia and western North America dominate the circulation. This pattern results in large scale meridional flow. Strong meridional flow into the Arctic, in particular, is often connected to blocking events (e.g., as analyzed by *Lejenäs and Okland*, [1983] and *Iversen and Joranger*, [1985]) particularly prevalent during winter. Note that by June the amplitude of the marked wintertime high

pressure regions have diminished, with a low now extending over northernmost Siberia and the Arctic Ocean. The summertime geopotential height pattern consists of features of much smaller scale and is no longer dominated by a pronounced wave number 2 pattern. As a result the large scale meridional flow from emission regions to the Arctic is reduced.

3.3 Episodic transport events into the Arctic during TOPSE

Consistent with the mean flow pattern, the frequency and strength of episodic transport events reaching the Arctic changes seasonally. While much of the CO in the Arctic region during winter has accumulated over a number of months from emission regions throughout the Northern Hemisphere [*Lamarque and Hess*, this issue], it is the relatively fast episodic transport of plumes from Europe and Siberia that contributes to the largest pollution events. Figure 5 gives the timeseries of CO over the North Pole for three months during the TOPSE campaign. The pollution events are predominantly due to transport of Eurasian emissions to the high Arctic (see section 4). During the TOPSE campaign the North Pole was more highly impacted by these episodic transport events than the region of the Canadian Arctic sampled during TOPSE. In fact, during the TOPSE campaign penetrations of pollution episodes into the Canadian Arctic were relatively rare. Nevertheless, some events did reach the Canadian Arctic and these are analyzed near the end of this section.

Figure 5 clearly shows that the overall CO concentration in the polar region decreases significantly from winter to summer (note the change in scales). This figure also indicates important qualitative differences in how pollution is transported over the pole during the different seasons. During winter and extending into spring, the plumes of pollutants reaching this remote location occur mostly in the lowest 300mb; during late spring and summer the surface values are generally low and the pollutant plumes occur in the midtroposphere. Additional confirmation that the resulting difference is indeed due to transport is given by the timeseries of Radon 222 (Figure 6). The emissions of radon in the model were set to be constant and uniform over land free of permanent ice ($1\text{atom}/\text{cm}^2/\text{sec}$ as in *Heimann et al.*, 1990, unaffected by snow cover). Rn^{222} decays radioactively with a loss rate of approximately 5.5 days. Any difference in the concentration of this species between seasons is purely due to differences in transport. Radon concentrations in the plumes are much higher in February than June, indicating the more efficient transport of this short-lived species to the Arctic. The largest plumes occur just above the surface in the winter. During summer the radon plumes extend higher to higher altitudes indicating faster transport to the middle troposphere during the summer months. Differences in the location of the CO and radon plumes are the result of differences in the location of the emissions and the short radon lifetime. Note that the radon plumes tend to occur at lower altitudes than those of CO.

While the same transport event is usually responsible for the transport of both plumes to the Arctic, the CO is emitted further south and, as argued in the next section, will consequently be lifted to a higher level due to the characteristic slope of the isentropic surfaces.

One of the largest transport events polluting the lower tropospheric Arctic took place between February 5 and February 15 (figure 7). It can be attributed to a very strong blocking event over the Pacific with the blocking high centered over Alaska and a strong wave number 2 pattern. This blocking pattern extended throughout most of the depth of the troposphere. This situation allows for efficient poleward transport of the pollution accumulated over the northern coast of Russia. This particular blocking event lasted for more than a week (February 5-15) and caused significant amount of pollution from Eurasia to be forced deep into the Canadian Arctic region (figure 7b). The details of the transport mechanism during this event are examined below.

4 Quantification of the differences in transport with the help of diagnostic tracers

In order to isolate the role of transport from that of other processes (e.g. chemistry, wet removal), and to obtain additional more quantitative information on the pathways and mechanisms of transport of pollutants, the model was rerun with several diagnostic tracers. We interpret these runs using an isentropic framework. While seasonal changes in meteorology are often complex and poorly understood, the isentropic perspective provides a simple explanation of how differences in seasonal and regional rates of heating and cooling lead to seasonal differences in transport. Results shown in sections 4.2, 4.3 and 4.4 show how these different rates of cooling and heating affect tracer transport in support of the isentropic interpretation. Before analyzing the simulations using the diagnostic trace species, we discuss our interpretation of the transport from the isentropic perspective.

4.1 Interpretation of the transport from an isentropic perspective

The isentropic perspective is based on the principle that in the absence of diabatic heating or cooling a parcel will simply be transported along a constant isentropic surface. Transport across the surface necessarily implies atmospheric heating or cooling, which can be achieved by latent heat release, transport of sensible heat, or the absorption and emission of long wave and short wave radiation. Therefore, knowledge of heating or cooling rates and the geography of isentropic surfaces provides a great deal of information about transport pathways. When diabatic heating (or cooling) and chemistry are slow compared with transport timescales, mixing will tend to homogenize tracer concentrations along isentropic surfaces and the isolines of the pollutant and those of the isentropic surfaces will be in close correspondence. In this case the concentrations of emitted pollutants (e.g., CO) will be highest on isentropic

surfaces at the same potential temperature as the pollutant's emissions. We show below that these assumptions are often met for trace species with a long lifetime, particularly during the winter months (see Figure 12).

Strong diabatic heating (leading to rapid cross isentropic transport) can be expected to occur in the boundary layer where the earth's surface provides a large heat source, and in regions of strong precipitation where large amounts of latent heat are liberated. These regions of strong heating are mostly balanced by the slow radiational cooling, which prevails throughout much of the free troposphere. Significant winter time cooling can also take place due to contact with cold land surface. Figure 8 shows the net diabatic rates from the NCEP analysis approximately 50mb above the surface for February 7 and 8, during the time period when the transport from Eurasia to the Arctic (section 3.3) is particularly strong. This figure shows cooling rates in the boundary layer over much of Siberia and North American of several degrees per day, due mostly to contact with the cold surface. Figure 9 gives the vertical heating profile for the Siberian region at this time. Strong cooling is indeed concentrated in the boundary layer. Above the boundary layer the cooling rates are on the order of 1K/day, as expected throughout most of the free troposphere. Regions with strong surface cooling are also characterized by persistent temperature inversions, particularly strong over parts of Eurasia and the high Arctic as shown in figure 10.

Transport from Eurasia to the lower arctic troposphere requires two conditions, namely: 1) strong diabatic cooling required for the transport of pollutants between their emission potential temperatures and the colder arctic temperatures, and 2) a temperature inversion that traps pollutants in the boundary layer limiting their mixing into the free troposphere and exposing them to the strong surface cooling. These two conditions are, as shown above, often met for the Eurasian emissions during winter, but are not met during the summer months, or for North American pollution, which because of the predominant track of weather systems is largely swept off the North American continent and over the North Atlantic. Figure 8 and the regionally averaged heating profile in figure 9 show considerable heating in the North Atlantic boundary layer during the winter months. This heating is predominantly due to sensible heating. Strong heating is to be expected during the winter months when cold continental air is advected over the warmer ocean waters. In fact the warm ocean temperatures and the strong vertical temperature gradient (figure 10) often cause the oceanic boundary layer to be unstable off North America during winter. The vertical profile of CO to the east of the primary European and American emission sources is shown in Figure 11. Over Eurasia considerable amount of CO is trapped in the boundary layer, while over the Atlantic the vertical gradient of CO is fairly flat, reflecting the strong mixing in the boundary layer and the rapid transport of pollutants to the free troposphere. These differences, as will be shown in more detail below, cause the transport of North American emissions to be very different from Eurasian emissions. The North American emissions, as they are swept out over the ocean are expected to be transported to higher potential temperature

surfaces. This, in connection with the fact that the North American emissions are often situated south of the polar front, makes it more probable that North American pollution will be initially transported to higher altitudes than pollutants from Eurasia. Consequently North American pollutants are less likely to impact the lower Arctic. Pollutant transport from Eurasia into the lower arctic troposphere occurs due to the rapid cooling of the pollutants as they move north over the cold snow covered surface of northern Europe and Asia under a strong temperature inversion. The European pollutants are less likely to be rapidly mixed into the free troposphere.

Different behavior is expected in summer. The Eurasian region experiences on average weak heating (not shown) during the summer months. The efficient winter cooling mechanism described above is absent. In addition, the boundary layer is less stable during summer as a result of the relatively high surface temperatures. This leads to fewer temperature inversions and faster vertical mixing of pollutants into the free troposphere. The summertime situation over North America is also different, with the land surface now generally warmer than that of the ocean. *Angevine et al.* [1996a, 1996b] discuss different scenarios for the transport of North American pollutants over the ocean during summer. The cooler ocean surface can result in a stable layer of high pollutant concentrations over the ocean, or in the upward transport of the pollution over a relatively clean marine layer.

4.2 Seasonal differences in the transport of the non-reactive and 7-day tracers

In the remainder of this section, we interpret the transport using two diagnostic tracers: one with a 7-day decay time (TR7, to diagnose the direct transport of short-lived species from their emission source) and one entirely non-reactive tracer (TRN, to diagnose transport behavior in the longer term). Both of these species are assigned the same source, which, in order to compare results for different months, is constant in time. The source is equal to the CO emission source in December. This emission footprint is characteristic of most anthropogenic pollutants, so that the results will be generally applicable. The model simulations are carried out for three periods corresponding to winter, spring and summer. Each of these periods is two months long: 1) December and January, 2) March and April, and 3) June and July. The first month from each of these periods is used for initialization. The results shown below are taken only from the second month. Note, that the seasonal differences in the distribution of the diagnostic tracers result only from differences in transport and not from the differences in chemistry or emissions. For all simulations the initial conditions and the boundary conditions are set to zero. The surface deposition is also set to zero.

The distributions of the non-reactive tracer (TRN) for January, April and July are shown in Figure 12, where the mixing ratio of TRN is monthly and zonally averaged. As expected from the discussion in sections 3.2-3.3 and 4.1, the concentration of this tracer throughout the lower part of the high latitude troposphere is highest in winter and decreases

with time. As shown in figure 13, the mixing ratios in this region decrease by more than a factor of two between January and July due to transport differences alone. On the other hand, TRN is highest in the summertime in the mid and upper troposphere south of about 60N, reflecting the more efficient vertical lifting of the pollution during the summer months.

Figure 12 also depicts the zonally and monthly averaged isentropes (solid black lines). In general, during all seasons, the isentropes slope upwards towards the pole due to the meridional gradient in temperature. In January, and for the most part in April, despite the averaging that encompasses various meteorological conditions, the isolines of the tracer and the isentropes tend to be parallel in the free troposphere. This indicates that rapid mixing and/or transport are taking place largely on surfaces of constant potential temperature, suggesting the value of an isentropic framework. Hess [2001] found similar results over the Pacific Ocean during outflow from Asia.

Figure 12 also demonstrates, however, that there also are large departures from isentropic transport during all seasons. Moreover, the location of the diabatic heating (cooling) and the subsequent transport change from winter to summer. During the winter months in the arctic latitudes the highest concentrations of TRN occur near the surface, and therefore at lower potential temperatures than the potential temperatures of the midlatitude emission regions. This implies that strong diabatic cooling must be taking place in the model during transport into this region, consistent with the discussion in section 4.1. During spring and summer the vertical gradient of TRN over the lower Arctic is reversed (i.e., TRN increases with height) and the highest concentrations of TRN are in the mid-troposphere at potential temperatures corresponding to those of the emission regions. This indicates that the cooling associated with transport from the emission regions to the Arctic is weaker than during the winter months and the transport more nearly isentropic. Note that during summer the vertical lofting over the emission regions increases while the overall transport into the high latitudes is weaker than in January. This change is consistent with an increase in convection and boundary layer mixing during the summer months. April represents an intermediate case, with weaker cooling than during winter and weaker heating than in summer.

The vertical lifting of TRN and its transport into the high latitudes in January and July is given quantitatively in figure 14. In agreement with the qualitative analysis presented above, the tracer is shifted to higher latitudes during winter (figure 14a). Figure 14b shows that significantly more of the tracer is contained in the lowest kilometer above the ground in winter than in summer at all latitudes and the ratio of winter to summer concentrations increases with latitude. A similar result applies in the 1-3 km altitude band. The opposite is the case above 6 km, where the summertime concentrations are higher at all latitudes consistent with a higher rate of vertical mixing (figure 14c).

Figures 12 through 14 reflect the long-term transport of TRN after it has traveled a long distance from its emission region. To examine the transport in the short term we examine the behavior of the diagnostic species with the seven day lifetime (TR7) (figure 15). Figure 15 should be compared with figure 12. The concentration of TR7 is much lower than TRN reflecting its short lifetime. The short lifetime implies a relatively small radius of influence away from its emission regions and acts to significantly lower the background concentrations of TR7. While the overall transport characteristics of this short lived trace species are rather similar to the inert tracer, important differences remain. First, note that the tracer isolines of TR7 do not follow isentropic surfaces to the same extent as those of TRN, suggesting that the timescale for mean meridional mixing on isentropic surfaces is longer than 7 days. Secondly, the ratio of winter to summer concentrations of TR7 in the high latitudes is significantly greater than that of TRN (compare figure 16 with figure 13). This is consistent with the TOPSE measurements (Figure 1), which show that the short lived hydrocarbons have higher amplitude seasonal cycles in the high latitudes than the longer-lived ones. We conclude that the seasonal differences in the short-term transport pathways to the Arctic are especially large.

4.3 Quantification of the diabatic heating and cooling during transport into the Arctic

In order to gain more quantitative information on the effects of diabatic heating, we labeled the non-reactive and 7-day tracers by the potential temperature at which they are emitted. The labeling is done in 10K intervals. If, for example, the potential temperature at the surface of a given grid box is 275K, only the tracer labeled between 270 and 280K is emitted. Since the surface temperature changes every 3 hours in our model, the emitted species are also updated with this frequency. With diabatic heating or cooling set to zero these tracers would be confined to the potential temperatures at which they are emitted, but since these rates are non-zero in the model, these tracers are redistributed to other potential temperatures.

Figure 17 clearly demonstrates the effects of heating and cooling on the distribution of the emitted tracer. This figure shows the zonally and monthly averaged mixing ratio of TRN labeled by the dominant emission temperature in winter (270-280K) and summer (290-300K). In these plots potential temperature is adopted as the vertical coordinate. Note that the potential temperature at the surface increases as one moves south. The emissions of the tracer are mostly over land and, therefore, within any particular latitude band the winter time emissions tend to be at relatively cold temperatures (the oceans, without significant emissions are at relatively higher temperatures). As a consequence, the highest concentrations are at the surface of the potential temperature plot. However, in summer, the emissions over land will be at relatively high potential temperatures (land surfaces are warmer than the ocean surfaces during summer) and will appear to be elevated from this zonally averaged isentropic perspective. Figure 17 shows that in both seasons a

large fraction of TRN is redistributed to other potential temperatures within the two months of integration (see also table 1). However, note the qualitative difference between the seasons: during winter significantly more of the emitted species sinks to colder temperatures due to strong diabatic cooling, particularly evident in the high latitudes; in the summer the sinking is less important. Therefore, during winter there are substantial concentrations of tracer present at potential temperatures much lower than their emitted temperature. This effect is shown quantitatively in table 1 for all latitudes and table 2 for high latitudes. In general, cooling to lower temperatures is favored in winter. Heating to higher temperatures can occur in both seasons but is stronger in summer. These differences are especially pronounced for the tracer transported into high latitudes (table 2). Note also, the differences in the tracer isolines between the two seasons indicating seasonal differences in adiabatic heating and cooling rates. In winter the isolines are flatter; in summer they are more bowed.

To gain additional insight into the seasonal differences between emission regions we examined differences in the wintertime transport of the North American and European emissions to the Arctic (Figure 18). In these simulations the tracer TR7 was labeled both by the potential temperature and its region of emission (N. American or European). The tracer TR7 allows an examination of the immediate transport pathways downwind from the emissions, rather than the transport along global pathways (as for TRN), which allow for successive cooling and heating of the airmasses as they are transported around the globe. Figure 18 shows the results for emissions in the 270-280K temperature range (corresponding to the largest emission temperature over both source regions). Clearly there is a qualitative difference between the transport of the North American and European emissions in the high latitudes. In the case of North American emissions the pollution is largely transported to higher levels, while the pollutants from Europe exhibit significant sinking. The percentage of tracer mass north of 70N within the 270-280K temperature bin, at temperatures above this bin, and at temperatures below it, is given in table 3. The difference between the two emission regions is large: over 45% of the European emissions between 270 and 280 K sink beneath 270K during the transport into the Arctic, compared with only 18% for the N. American emissions. In the case of the North American emission, on the other hand, nearly 60% is lifted above 280K compared to only 18% for the European source. During summer both the European and N. American sources tend to be lifted to higher potential temperatures (figure not shown).

The above analysis demonstrates important differences between wintertime transport from Europe and North America consistent with the hypothesis outlined in section 4.1. These differences can be explained largely by examining the conditions downwind from the emissions. In winter, the relatively warm Atlantic ocean is immediately downwind of the large emissions over the east coast of the U.S. Cold continental air swept over the Atlantic is rapidly warmed to higher potential temperatures, often in strong convective mixing events. Secondly a strong storm track exists

off the east coast of the U.S. with its associated diabatic heating [Hoskins or Wallace]. Similar behavior governs the transport of emissions in the Eastern Asia (not shown). This contrasts to the situation over Europe, where no strong storm track exists and the emissions are often swept over a cold land surface capped by an inversion. During summer there is more vertical mixing in the boundary layer over all emission regions due to the strong surface heating. Thus, the pollution plumes tend to be lifted from the boundary layer from the onset. In addition, deep convection acts to further redistribute the pollution in the vertical. The cooling mechanism, whereby European pollutants are transported over a cold surface under a strong inversion does not exist during the summer months.

4.4 Quantification of the contribution of main regional sources to the transport into the Arctic

The last series of simulations discussed in this paper is designed to quantify the contribution of the major emission regions to the overall concentrations of pollutants in the middle and high latitudes of the Northern Hemisphere. Here, we tag TRN and TR7 by one of five different emission regions: 1) North America, 2) Eurasia west of 40E (central and western Europe, hereafter referred to as the European source), 3) Eurasia east of 40N and north of 45N (predominantly emissions from Siberia), 4) Eurasia east of 40N and south of 45N (predominantly emissions from the Far East), and 5) global emissions north of 45N. Eurasia was split into 3 regions to differentiate between the large emissions in central and Western Europe, the sparse but northern emission source over Siberia, and the high emissions located over China, Korea and Japan. For these simulations the tracers were not differentiated by the potential temperature of the emissions. The latitudinal distribution of the emissions in each of these regions is given in Table 4.

4.4.1 Winter Case

Figure 19a shows the zonally and monthly averaged concentration of TRN in January, and the contribution from each of the 5 emission regions to this averaged concentration. In the lowest arctic atmosphere the largest contribution to TRN is from the European source in agreement with the previous discussion. The North American contribution is small (less than 15%) there, given the strength of this source (table 4). The contribution from the North American emissions increases with height, however, and becomes the dominant contributor in the upper troposphere (again consistent with the previous discussion). The SE Asian sources of TRN south of 45N, although large, contribute relatively little to the overall concentrations of TRN in the mid- and high- latitudes. These emissions occur at relatively high potential temperature and experience rapid mixing when transported over the Pacific, similar to the behavior of the North American emissions when transported over the Atlantic. However, while the N. American emissions are an important contributor to the upper arctic troposphere, the impact of the SE Asian emissions is relatively small. The prevailing

circulation moves these emissions south, away from the Arctic, and eventually out of the model domain. The Siberian emissions, despite their small magnitude (only 7% of the total source), contribute more than 20% to the TRN budget in the lower Arctic. They are ideally located on the pathway of transport to the Arctic and they are emitted at low potential temperatures. The pollution emitted north of 45N contributes to over 50% of the pollution in the high latitude lower troposphere, but to less than 50% of the pollution above 5 km. Due to the characteristic slope of isentropes in the troposphere (Figure 12) the more southerly emissions are more easily transported (i.e., without significant cooling or heating) into the upper troposphere than those to the north.

An analogous figure to 19a, but for TR7 is shown in Figure 19b. One of the main differences between the distribution of TRN and TR7 is the larger fraction of North American emissions in the high troposphere over the Arctic in the case of TR7. In accordance with our previous discussion, this again suggests the N. American emissions are lifted faster to higher heights (they are emitted at higher potential temperatures than the European emissions and are immediately subject to more diabatic heating). In the lowest part of the arctic atmosphere the contribution of the North American emissions to TR7 is even smaller than that to TRN, again indicating the lack of direct transport. The Siberian sources of TR7, due to their proximity to the Arctic, play a significant role comparable to that of the European emissions. Emissions of TR7 north of 45N contribute more near the surface than TRN and less aloft. Lifting this tracer aloft requires diabatic heating, which is slow compared to its short lifetime.

The plots above represent zonal averages. It is interesting to analyze a comparable plots for TR7 (figure 20) averaged only for the longitudes over which TOPSE measurements took place (105W-60W). One of the most striking features is the strong transport barrier for N. American emissions between 50°N and 60°N that extends from the surface up to the mid-troposphere. This barrier corresponds roughly to the location of the arctic front during winter. Despite the large geographical distance, the European emissions still make the largest contribution to the near surface pollution in the Canadian Arctic. Note that this analysis is based on monthly means. The contribution of a given region during a specific transport event can be naturally much higher.

4.4.2 Summer Case

In section 4.3 it was shown that in July both the European and North American sources can be lifted to higher potential temperature surfaces and therefore to higher altitudes than during January. The impact of this can be seen for TR7 (figure 21, compare with figure 19b) and to a lesser degree for TRN (not shown). During July the European emissions, due to their northerly location, are the main contributor to TR7 throughout the troposphere north of 50N. This is true throughout the depth of the troposphere, and not only in the lower troposphere as is the case in January. In

general, there is less vertical gradient in the contribution of the various regions during summer, suggesting greater vertical mixing for all sources. As shown in figure 12, there is less overall transport to the lower Arctic in July than in January. However, even in summer the European emissions are still the dominant contributor to pollutants in the lower arctic troposphere. This is especially true for the short lived tracer due to the relative proximity of Europe to the Arctic. In the TOPSE region during summer there is still an effective transport barrier (not shown) similar to the one in figure 20 for North American emissions, but now located further north, between 60 and 70N.

5 Conclusions

In this study we use a regional chemical transport model to examine the seasonal differences in the transport of midlatitude pollution. After demonstrating that the model reproduces the measured seasonality of a number of the measured hydrocarbons during the TOPSE campaign, we analyze the differences in model transport between seasons and between locations. To separate the effect of chemistry from that of transport we simulated the transport of diagnostic tracers which were either non-reactive or had a constant decay lifetime of 7 days. Simulations conducted with these tracers show that due to transport differences alone, there is a large seasonal cycle in the concentrations of tracers in the high latitudes with the largest seasonal differences in the lowest few hundred milibars above the surface. The seasonal cycle is larger for the short lived tracer.

This paper quantifies the contributions of 5 emission regions to the concentrations of the non-reactive and 7-day tracer in the mid and high latitudes of the N. Hemisphere and over the particular sector corresponding to the TOPSE measurements. During winter the European source is clearly dominant in the lower half of the Arctic. This is consistent with studies of the anthropogenic trace elements in Arctic Haze [e.g. *Lowenthal and Rahn*, 1985], studies of the meteorology over the region [e.g., *Raatz*, 1991], and previous modeling [*Levy and Moxim*, 1989]. Budgets for the longitudes sampled during the TOPSE campaign (105W to 60W) show a strong mixing barrier to North American emissions in the lower troposphere at approximately 60N. South of this line, and particularly below 4km, the simulations suggest that during TOPSE campaign predominately sampled North American pollution. North of this barrier the contribution of the North American emissions is generally small near the surface, but increases with height. This is especially true for shorter lived tracers. During summer the pollution is better mixed in the vertical and the strong mixing barrier over N. America moves considerably north, presumably following the location of the arctic front. The relative contributions are likely to change for species with different lifetimes, for species with different emission patterns, and for species affected by wet removal and presence of secondary sources (e.g. Arctic Haze).

Examining the transport from an isentropic perspective provides a clear and coherent picture of the transport differences between the winter and summer months and transport from different emission regions. While some of the differences are due to large scale seasonal changes in the tropospheric circulation described in section 3.2, the simple isentropic perspective given here goes a long way towards explaining the seasonal transport differences and the predominant transport pathways from the various emission regions. During winter, the European emissions are the main polluter of the lower tropospheric Arctic, due to the combination of the favorable meteorological conditions, the presence of the cold, stable boundary layer over Eastern Europe and Siberia, and the geographic proximity of the emissions to the Arctic, with the emission sources often located north of the polar front. With the help of the diagnostic tracers labeled by the potential temperature of the emissions we show that the European emissions in winter often experience strong diabatic cooling as they are transported northwards. This transport pattern differs considerably from that of the North American (and also South East Asian) emissions. Instead of being transported in stable continental air, the pollution plumes from North America (and Asia) are transported over the relatively warm ocean. Strong vertical mixing occurs as cold continental air is transported over the warmer oceanic surface. However, in addition a prominent storm track located off the east coast of the U.S. and Asia provides an additional lifting mechanism (note, that an analogous storm track does not occur to the east of Europe). The release of latent heat associated with the frequent precipitation events within the storm track, further transports pollutants upward across isentropic surfaces. This strong diabatic lifting helps to make the North American emissions the main contributor to the upper tropospheric budget in the Arctic. The other contributing factor is that North American emissions are generally located south of the European emissions (often at higher surface temperatures). Therefore, even adiabatic transport (see Figure 12) will tend to transport North American emissions to higher heights than their European counterparts.

Similar isentropic arguments can be invoked to explain the seasonal differences in transport. During summer the pollution is more likely to be diabatically mixed to higher levels over all the emission regions due to greater boundary layer and convective mixing. As a result, the contribution of the various emission regions remains relatively constant with height. The largest seasonal differences are simulated in the lower tropospheric Arctic. These differences are partially due to the large scale circulation which is less conducive to strong meridional transport during the summer months, but also to the absence of an efficient cooling mechanism during summer which allows the pollution plumes to rapidly cool diabatically as they are transported to the north. On the contrary, during summer the emissions are more apt to be transported upwards by the greater heating. Furthermore, in summer, the majority of the emissions are located south of the polar front so there is an additional isentropic barrier which they must cross to reach the lower arctic

troposphere. As a consequence, during summer emissions are transported to the Arctic at higher vertical levels, leading to a positive tracer gradient with height in the lower arctic troposphere.

APPENDIX

The goal of this appendix is to describe the major differences between the model used in this study and its earlier version described in *Hess et al.*, 2000. Most of the changes were introduced to make the model suitable for the high latitudes – the model from *Hess et al.*, 2000 was used to simulate the conditions present over the subtropical Pacific. The changes are itemized below.

- 1) Daily sea-ice was included in MM5 based on satellite observations archived at the National Snow And Ice Data Center [*Cavalieri et al.*, 2000, *Comiso*, 2000] by changing the surface type from water to ice when the observed coverage exceeded 50%. This modification allowed the surface temperatures to reach sub-zero values and allowed differences in transport to be simulated over water and ice surfaces.
- 2) A moisture scheme allowing for the formation of snow, ice and supercooled water was chosen in MM5 [*Grell et al.*, 1993a]
- 3) The parameterization of wet removal has been expanded to include the removal of species frozen in ice and the removal of nitric acid that deposits on the surface of ice (through snow fallout). As in *Hess et al.*, [2000] the *Giorgi and Chameidies* [1985] scheme is used for the partitioning, but now instead of partitioning simply between the gaseous and aqueous phase as in *Hess et al.*, [2000], the species is partitioned between the aqueous, ice and gas phase. The removal of a species due to wet deposition (R) can be written as:

$$R = \frac{\chi}{10^6 \cdot (L_{aq} + L_{ice}) + 1/(HRT)} \cdot (P_{aq} + P_{ice})$$

where χ is the mixing ratio of the removed tracer, L_{aq} is the liquid content of a cloud including rain water and cloud water, L_{ice} is the ice content of a cloud including ice and snow, H is Henry's Law constant, R is the gas constant, T the temperature and P_{aq} and P_{ice} the removal efficiency of rain and snow ($\text{kg}/\text{m}^3/\text{sec}$). Here we assume that a species is incorporated into liquid before it freezes into snow (or ice) and remains in the snow and ice after it freezes. The removal efficiency of rain water from a given layer is approximated as:

$$P_{aq}(z) = \frac{q_{rain}(z) \cdot \rho_{air}(z) \cdot \delta_{cloud}}{\int_0^{z_{top}} (q_{rain}(z) + q_{snow}(z)) \cdot \rho_{air}(z) \cdot \delta_{cloud} \cdot dz} \times P_r$$

A similar equation applies for P_{ice} , with q_{rain} replace by q_{snow} in the numerator. Here P_r is the precipitation rate at the surface (sum of rain and snow) and q_{rain} and q_{snow} are the mixing ratio of rain and snow in a cloud (kg/kg). The δ_{cloud} parameter, defined to be 0.1 outside of a cloud and 1 within a cloud, is added to account for higher scavenging of gases in a cloud than below clouds

In addition, the deposition of nitric acid on ice surface was added to account for the suggested fast deposition of HNO₃ on ice [e.g. *Abbatt 1997, Zondlo et al., 1997*]. The number of molecules of HNO₃ removed per unit volume from the gas phase due to deposition on ice surface can be defined as: $\chi_{ice} = A_{ice} \cdot D$, where A_{ice} is the surface area of ice crystals per unit volume, and D is the number of sites at which HNO₃ can stick to ice surface per cm² of ice. D is assumed here to be 2×10^{14} molecules/cm² (after *Abbatt, [1997]*). Making the assumption that ice crystals are spherical, A_{ice} can be obtained from the empirical relationship between ice cross sectional area (S_{ice}) and ice water content (q_{ice}) which we derived by fitting data from the ARM campaign (*Heymsfield, private communication*) using the functional form from *Heymsfield and McFarquahar [1996]*:

$$A_{ice} = 4 \cdot S_{ice} = 4 \cdot (50.4 \cdot (q_{ice})^{0.872})$$

The amount removed from a model layer by deposition onto ice is then assumed to be

$$A_{ice} \cdot D \cdot \frac{P_{ice}}{(q_{ice} + q_{snow}) \cdot \rho_{air}}$$

where the ratio estimates the rate of removal of snow from a layer.

4) The reaction accounting for the heterogeneous removal of N₂O₅ on the surface of aerosols was added. The rate of removal (1/sec) is then simply given by: $\text{gam} \cdot S \cdot \sqrt{8kT/(\pi \cdot M)}$, where gam is the reaction probability, S is the surface area of the aerosols calculated from the equilibrium size distribution, T is temperature and molecular weight of N₂O₅. The monthly averaged distribution of aerosol mixing ratio was taken from *Barth et al., [1999]*. The surface area of aerosols was calculated assuming a log-normal size distribution of dry aerosols with a mean radius of .05 μm and a standard deviation of 2. At a given relative humidity a new equilibrium size distribution was calculated using the method given in *Kiehl et al, [2000]* assuming the aerosol is composed of (NH₄)₂SO₄. We assumed efflorescence occurs at 35% relative humidity regardless of temperature. We used a temperature dependence of gam similar to that given in *Hallquist et al., [2000]*: $\text{gam} = \text{gam}_{300} + (\text{gam}_{200} - \text{gam}_{300}) \cdot (300 - t) / 100$, where gam_{300} and gam_{200} are rate coefficients at 300 and 200 K, respectively, where they are set equal .003 and .0185. These rates are lower than those recommended by *Hallquist et al., [2000]*. Since the total removal rate is equal to the surface area times the reaction

probability and because the surface area is not well know, these rates were lowered to increase NO_x in HANK to bring it into better agreement with the measurements.

5) A lightning source of NO_x was added based on the parameterization of *Price and Rind* [1992]. The lightning source of NO_x was distributed in the vertical according to the recommendations of *Pickering et al.*, [1998].

6) Ozone was relaxed in the stratosphere above PV=2 surface with a relaxation time of 5 days to values determined by the O₃/PV parameterization described in *Klonecki et al.* (manuscript in preparation). The parameterization consists of quadratic fits developed with the help of the ozone sonde data and matching PV values from NCEP analysis. The fits change monthly but a single fitting function describes the O₃-PV dependence throughout the lower stratosphere for the whole model domain. The stratospheric values of HNO₃ were similarly relaxed based on the measured O₃/HNO₃ ratio from the TOPSE campaign.

7) The chemistry mechanism was updated following the recommendations from *Sander et al.*, [2000]. In addition the following changes were introduced:

-oxidation scheme of C₂H₄ was expanded following the mechanism from *Niki et al.*, [1981].

-The rate of the reaction of the lumped organic nitrate was increased following *Brasseur et al.*, [1998] (MOZART model). In addition, the washout rate of nitrates was set to four times that of H₂O₂ following MOZART (....., personal communication) and yields of a number of organic reactions were changed. The full mechanism can be found on the WWW at :

8) The emissions used in the model are based on the 1x1 inventory developed by Horowitz et al.,

References:

Abbatt, J. P. D., Interaction of HNO₃ with water-ice surfaces at temperatures of the free troposphere, *Geophys. Res. Lett.*, 24, 479-1482, 1997.

Angevine, W. M., M. Trainer, S. A. McKeen, and C. M. Berkowitz, Mesoscale meteorology of the New England coast, Gulf of Maine, and Nova Scotia: Overview, *J. Geoph. Res.*, 101, 28893-28901, 1996.

- Angevine W. M., M. P. Buhr, J. S. Holloway, M. Trainer, D. D. Parrish, J. I. MacPherson, G. L. Kok, R. D. Schillawski, and D. H. Bowlby, Local meteorological features affecting chemical measurements at a North Atlantic coastal site, *J. Geoph. Res.*, 101, 28935-28946, 1996.
- Atlas, E. et al., The TOPSE experiment: introduction, this issue.
- Barrie L. A. and R. M. Hoff, Five years of air chemistry observations in the Canadian Arctic, *Atmosph. Environ.*, 19, 1995-2010, 1985.
- Barie, L. A., Arctic Air Pollution: An overview of current knowledge, *Atmos. Environ.*, 20, 643-663, 1986.
- Barie L. A., J. W. Bottenheim, and W. R. Hart, Polar Sunrise Experiment 1992 (PDSE 1992): Preface, *J. Geoph. Res.*, 99, 25,313-25,314, 1994.
- Barth, M. C., P. J. Rasch, J. T. Kiehl, C. M. Benkovitz, and S. E. Schwartz, Sulfur chemistry in the National Center for Atmospheric Research Community Climate Model: Description, evaluation, features and sensitivity to aqueous chemistry, *J. Geoph. Res.*, 105, 1387-1415.
- Blake, N.J., D. R. Blake, B. C. Sive, A. S. Katzenstein, S. Meinardi, O. W. Wingenter, and F. S. Rowland, The seasonal evolution of NMHCs, light alkyl nitrates and selected halocarbons at mid to high northern latitudes during TOPSE, *J. Geophys. Res.*, this issue.
- Blake, D.R. and S. Rowland, Global atmospheric concentration and source strength of ethane, *Nature*, 321, 231, 1986.
- Brasseur G.P., D. A. Hauglustaine, S. Walters, P. J. Rasch, J.F. Muller, C. Granier, XX Tie, MOZART, a global chemical transport model for ozone and related chemical tracers 1. Model description, *J. Geoph. Res.*, 103, 28265-28289, 1998.
- Carlson, T.N, Speculations on the movement of polluted air to the Arctic, *Atmos. Environ.*, 15, 1473-1477, 1981.
- Cavalieri, D., P. Gloerson, and J. Zwally. 1990 - March 2000,. DMSP SSM/I daily polar gridded sea ice concentrations. Edited by J. Maslanik and J. Stroeve. Boulder, CO: National Snow and Ice Data Center, Digital media.
- Comiso, J. 1990 - March 2000. DMSPSSM/I daily polar gridded sea ice concentrations. Edited by J. Maslanik and J. Stroeve. Boulder, CO: National Snow and Ice Data Center, Digital media.
- Cziczo, D. J. and J. P. D. Abbatt, Deliquescence, efflorescence, and supercooling of ammonium sulfate aerosols at low temperature: Implications for cirrus cloud formation and aerosol phase in the atmosphere, *J. Geoph. Res.*, 104, 13,781-13,790, 1999.
- Giorgi, F. and W. L. Chameides, The rainout parameterization in a photochemical model, *J. Geoph. Res.*, 90, 7872-7880, 1985.

- Grell, G. A., J. Dudhia, D. R. Stauffer, A description of the fifth generation Penn State/NCAR Mesoscale Model (MM5) (MM5), NCAR/TN-398+IA, National Center for Atmospheric Research, Boulder, CO, 116pp, 1993.
- Grell, G. A., Prognostic evaluation of assumptions used by cumulus parameterizations, *Mon. Wea. Rev.*, 121, 764-787, 1993.
- Hallquist, M., D. J. Stewart, J. Baker, and R. A. Cox, Hydrolysis of N₂O₅ on Submicron Sulfuric Acid Aerosols, *J. Phys. Chem. A*, 104, 3984-3990, 2000.
- Heimann, M., P. Monfray, Georges P., Modeling the long-range transport of ²²²Rn to subantarctic and antarctic areas, *Tellus*, 42B, 83-99, 1990.
- Hess, P. G., Model and measurement analysis of springtime transport and chemistry of the Pacific basin, *J. Geophys. Res.*, 106 (D12): 12689-12717 JUN 27 2001
- Hess, P. G., S. Flocke, JF. Lamarque, M. C. Barth, S. Madronich, Episodic modeling of the chemical structure of the troposphere as revealed during the spring MLOPEX 2 intensive. *J. Geophys. Res.*, 105, 26809-26839, 2000.
- Heymsfield, A and G. M. Farquhar, High albedos of cirrus in the tropical Pacific warm pool: microphysical interpretations from CEPEX and from Kwajalein, Marshall Islands *J. Atmos. Sci.*, 53, 2424-2451, 1996.
- Iversen T. and E. Joranger, Arctic air pollution and large scale atmospheric flows, *Atmos. Environ.*, 19, 2099-2108, 1985.
- Jobson, B. T., H. Niki, Y. Yokouchi, J. Bottenheim, F. Hopper and R. Leitch, Measurements of C₂-C₆ hydrocarbons during the 1992 Polar Sunrise Experiment: Evidence for Cl-atom and Br-atom chemistry, *J. Geophys. Res.*, 99, 25,329-25,344, 1994.
- Kiehl, J. T., T. L. Schneider, P. J. Rasch and M. C. Barth, *J. Geophys. Res.*, 105, 1441-1457, 2000.
- Klonecki A. and P.G. Hess, Analysis of the correlation between O₃ and potential vorticity in the lower stratosphere and possible applications, manuscript in preparation.
- Lamarque J-F., P.G.Hess, Model analysis of the temporal and geographical origin of the CO distribution during the TOPSE campaign, this issue.
- Lejnas H. and H. Okland, Characteristics of northern hemisphere blocking as determined from a long time series of observational data, *Tellus*, 35A, 350-362, 1983.
- Levy, H. II and Moxim, W.J., Examining the global impact of local/regional air pollution: The role of global chemical transport models. In *Air Pollution Modeling and its Application VII*, Plenum Publishing Corp., 139-157, 1989
- Niki, H., P.D. Maker, C.M.Savage, and L.P. Breitenbach, An FRIR study of mechanisms for HO radical initiated oxidation of C₂H₄ in the presence of NO: detection of glycoaldehyde, *Chem. Phys. Lett.*, 80, 499-503.

- Penkett, S. A., N. J. Blake, P. Lightman, A. R. W. Marsh, and P. Anwyl, The seasonal variation of nonmethane hydrocarbons in the free troposphere over the North Atlantic Ocean: Possible evidence for extensive reaction of hydrocarbons with the nitrate radical, *J. Geophys. Res.*, 98, 2865-2885, 1993.
- Pickering, K. E., Y. Wang, W.-K. Tao, C. Price and J.-F. Muller, Vertical distributions of lightning NO_x for use in regional and global chemical transport models, *J. Geophys. Res.*, 31203-31216, 1998.
- Price, C. and D. Rind 1992: A simple lightning parameterization for calculating global lightning distributions, *J. Geophys. Res.*, 97, 9919-9933, 1992.
- Raatz, W. E., The climatology and meteorology of Arctic air pollution, *Pollution of the Arctic Atmosphere*, edited by W.T.Sturges, pp. 13-42, 1991.
- Raatz W. E. and G. E.Shaw, Long-range transport of pollution aerosols into the Alaskan Arctic, *J. Clim. Appl.Met.* 23, 1052-1064, 1984.
- Smolarkiewicz, P. K., A fully multidimensional positive definite advection algorithm with small implicit diffusion, *J. Comput. Phys.*, 54, 325-362, 1984.
- Solberg, S., C. Dye, and N. Schmidbauer, Carbonyls and nonmethane hydrocarbons at rural European sites from the Mediterranean to the Arctic, *J. Atmosp. Chem.*, 25, 33-66, 1996.
- Zondlo M. A., S. B.Barone, and M. A. Tolbert, Uptake of HNO₃ on ice under upper tropospheric conditions, *Geophys. Res. Lett.* 24, 1391-1394, 1997.

Captions for Tables and Figures

Table 1 Distribution of TRN labeled by emission potential temperature for January and July. The tracer can be either in, below, or above its emission potential temperature bin. The values are given for the dominant emissions temperature for both seasons and summed over all bins. The given values are monthly averages.

Table 2 As in table 1 but the mass of the tracers is integrated only for latitudes north of 70°N.

Table 3 Distribution of the TR7 labeled by the emission potential temperature and region of emission (Europe and N. America) for January. The mass of the tracer is integrated only for latitudes north of 70°N.

Table 4 Fractional contribution of regional sources to the overall tracer (CO) emissions (first column) and contribution of the regional sources to the overall source by latitude (remaining columns). NE= No Emissions for a given latitude band.

Figure 1 Comparison of the observed (solid line) and modeled (dashed line) median mixing ratios for a) ethene, b) ethane, c) propane, and d) CO for data sampled in the 1 to 6km altitude range. Modeled data is interpolated spatially and temporally to the location and time of each measurement. The error bars indicate the +/- 25 percentiles. The figures in the left column show data sampled between 58°N and 90°N binned by deployment. The figures in the right column show data binned by latitude for two deployments: one just before the winter-spring transition (open symbols) and one after (filled symbols).

Figure 2 Measured and modeled medians of propane for deployments 4,5,6,and 7. Data are sampled above 1km and north of 58°N and are binned by altitude.

Figure 3. Concentration of CO (ppbv) at 850mb from the simulation with 56 chemical species for February, April and June. White areas indicate regions where the surface pressure is lower than 850mb.

Figure 4. Monthly averaged geopotential heights (in meters) from MM5 at 850mb for February, April and June

Figure 5 Time series of CO (ppbv) over the North Pole for February, April and June

Figure 6 As in figure 5 but for $Rn^{222}(*10^{20})$

Figure 7 a) geopotential height at 850mb on February 8, 2000, and **b)** CO (ppbv) at 850mb on February 9, 2000

Figure 8. Diabatic heating rate from the NCEP model averaged for February 7 and 8 at approximately 50mb above the surface.

Figure 9 Vertical profile for the total adiabatic heating rate from the NCEP model (K/day) averaged for February 7 and 8 for the region over Siberia: 30E-120E and 50N-70N (solid line) and over the North Atlantic: 75W-60W, 30N-45N (dashed line).

Figure 10 Temperature difference between 1000 and 100m above the surface for February 7-8, 2000.

Figure 11 Vertical profile for CO (ppbv) averaged for February 7 and 8 for the regions described in caption for figure 9

Figure 12 Monthly and zonally averaged mixing ratios of TRN for January, April, and July in ppbv (solid lines). The dashed lines indicate the monthly and zonally averaged isentropes (K).

Figure 13 Ratio of the monthly and zonally averaged mixing ratios of the TRN for January and July

Figure 14 Zonally and vertically integrated mass of TRN averaged for the month of January and July. The vertical integration carried for a) the whole troposphere, b) from the surface to 1km and 1km to 3km, c) from 3km to 6km and from 6km to the top of the model

Figure 15 Monthly and zonally averaged mixing ratios of TR7 for January, April, and July in ppbv (solid lines). The dashed lines indicate the monthly and zonally averaged isentropes (K).

Figure 16 Ratio of the monthly and zonally averaged concentrations of TR7 for January and July

Figure 17 Monthly and zonally averaged concentration of TRN interpolated to surfaces of constant potential temperatures for a) the main emission temperature in January: 270-280K and b) the main emission temperature in July: 290-300K. More detailed description in the text. Due to surface temperature variations in longitude, all longitudinal points on a particular potential temperature surface may not be defined. A zonal average is taken only if the value at a given level is defined for at least 25% of all grids at that latitude for all time steps during the month, otherwise the point is left blank.

Figure 18 Monthly and zonally averaged concentration of TR7 emitted at potential temperature of 270-280K in January interpolated to surfaces of constant potential temperatures for a) the tracer emitted over Europe, and b) the tracer emitted over N. America

Figure 19a Monthly and zonally averaged concentrations of TRN for January (in ppbv) and the contribution of the 5 main emissions regions (in %)

Figure 19b As in Figure 19a but for TR7.

Figure 20 As in figure 19b but the average calculated only over the TOPSE region (105W-60W)

Figure 21 As in figure 19b but for July

Table 1 Distribution of TRN labeled by emission potential temperature for January and July. The tracer can be either in, below, or above its emission potential temperature bin. The values are given for the dominant emissions temperature for both seasons and summed over all bins. The given values are monthly averages.

MONTH	Tracer type (emitted for Θ between:)	Total mass of the tracer in the model (Tg)	% of the tracer in the original potential temperature layer	% of the tracer below the layer (cooling)	% of the tracer above the layer (heating)
JANUARY	270-280K	13.3	25.1	20.6	54.3
	Sum over all temperature bins	33.1	27.0	25.0	48.0
JULY	290-300K	13.1	26.0	6.9	67.1
	Sum over all temperatures bins	31.9	27.3	13.3	59.4

Table 2 As in table 1 but the mass of the tracers is integrated only for latitudes north of 70°N.

MONTH	Tracer type (emitted for Θ between:)	Total mass of the tracer (Tg) north of 70°N	% of the tracer in the original potential temperature layer	% of the tracer below the layer (cooling)	% of the tracer above the layer (heating)
JANUARY	270-280K	1.95	25.6	43.3	31.1
	Sum over all temperature bins	4.78	23.8	44.9	31.3
JULY	290-300K	1.54	23.8	23.5	52.7

	Sum over all temperature bins	3.33	22.5	25.2	52.3
--	-------------------------------	------	------	------	------

Table 3 Distribution of the TR7 labeled by the emission potential temperature and region of emission (Europe and N. America) for January. The mass of the tracer is integrated only for latitudes north of 70°N.

Region of emissions	Tracer type (emitted for Θ between:)	Total mass of the tracer (Tg) north of 70°N	% of the tracer in the original potential temperature layer	% of the tracer below the layer (cooling)	% of the tracer above the layer (heating)
Europe	270-280K	0.11	35.9	45.6	18.5
	Sum over all temperature bins	0.22	33.4	43.1	23.4
N. America	270-280K	0.04	23.4	17.8	58.8
	Sum over all temperature bins	0.11	21.8	21.6	56.6

Table 4 Fractional contribution of regional sources to the overall tracer (CO) emissions (first column) and contribution of the regional sources to the overall source by latitude (remaining columns). NE= No Emissions for a given latitude band.

Emission region	%of total source	South of 30N % of total	30N-40N % of total	40N-50N % of total	50N-60N % of total	60N-70N % of total	70N-80N % of total	80N-90N % of total
All regions	100	16.6	36.5	27.3	18.2	1.4	0.01	NE
N. America	24.9	1.4	15.5	7.5	0.4	0.02	0.0006	NE
Eurasia W of 40E (Europe)	30.4	NE	2.8	13.9	13.0	0.8	0.0009	NE
Eurasia S of 45, E of 40E	32.6	12.8	15.9	4.0	NE	NE	NE	NE
Eurasia N of 45, E of 40E	7.2	NE	NE	1.9	4.8	0.5	0.004	NE
North of 45N	33.7	NE	NE	14.2	18.2	1.4	0.01	NE