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D. T. Shindell, H. Teich, M. Chin, F. Dentener, R. M. Doherty, G. Faluvegi, A. M. Fiore, P. Hess, I. A. Mackenzie, M. G. Sanderson, et al.

► **To cite this version:**

D. T. Shindell, H. Teich, M. Chin, F. Dentener, R. M. Doherty, et al.. A multi-model assessment of pollution transport to the Arctic. *Atmospheric Chemistry and Physics Discussions*, 2008, 8 (3), pp.8385-8429. hal-00328325v1

**HAL Id: hal-00328325**

**<https://hal.science/hal-00328325v1>**

Submitted on 18 Jun 2008 (v1), last revised 9 Nov 2020 (v2)

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# A multi-model assessment of pollution transport to the Arctic

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Received: 28 March 2008 – Accepted: 1 April 2008 – Published: 6 May 2008

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Published by Copernicus Publications on behalf of the European Geosciences Union.

ACPD

8, 8385–8429, 2008

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## Abstract

We examine the response of Arctic gas and aerosol concentrations to perturbations in pollutant emissions from Europe, East and South Asia, and North America using results from a coordinated model intercomparison. These sensitivities to regional emissions (mixing ratio change per unit emission) vary widely across models and species. Intermodel differences are systematic, however, so that the relative importance of different regions is robust. North America contributes the most to Arctic ozone pollution. For aerosols and CO, European emissions dominate at the Arctic surface but Asian emissions become progressively more important with altitude, and are dominant in the upper troposphere. Sensitivities show strong seasonality: surface sensitivities typically maximize during boreal winter for European and during spring for East Asian and North American emissions. Mid-tropospheric sensitivities, however, nearly always maximize during spring or summer for all regions. Deposition of black carbon (BC) onto Greenland is most sensitive to North American emissions. North America and Europe each contribute ~40% of total BC deposition to Greenland, with ~20% from East Asia. Elsewhere in the Arctic, both sensitivity and total BC deposition are dominated by European emissions. Model diversity for aerosols is especially large, resulting primarily from differences in aerosol physics and removal. Comparison of aerosols with observations indicates problems in either the models or interpretation of the measurements. For gas phase pollutants such as CO and O<sub>3</sub>, which are relatively well-simulated, the processes contributing most to uncertainties depend on the source region. Uncertainties in the Arctic surface CO response to emissions perturbations are dominated by emissions for East Asian sources, while uncertainties in transport, emissions, and oxidation are comparable for European and North American sources. At higher levels, model-to-model variations in transport and oxidation are most important. Differences in photochemistry appear to play the largest role in the intermodel variations in Arctic ozone sensitivity.

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## 1 Introduction

Transport of pollution to the Arctic affects both air quality and climate change. While levels of pollutants such as tropospheric ozone and aerosols are generally lower in the Arctic than in industrialized areas, they can have substantial impacts on climate. For example, aerosols can greatly perturb the Arctic radiation balance (Garrett and Zhao, 2006; Lubin and Vogelmann, 2006). Though pollutant levels outside the Arctic may in fact have a larger influence than local pollutant levels on Arctic climate (Shindell, 2007), it is important to understand the sources of the pollution that reaches the Arctic. This pollution alters local radiative fluxes, temperature profiles and cloud properties. Pollutant levels within the Arctic are especially important for climate in the case of black carbon (BC), which clearly has a strong local climate impact when it is deposited onto snow and ice surfaces, reducing their albedo (Hansen and Nazarenko, 2004; Jacobson, 2004; Warren and Wiscombe, 1980; Vogelmann et al., 1988).

While air pollution in most heavily populated areas of the world comes predominantly from local and regional emissions, pollution in the remote Arctic is primarily a result of long-range transport from source regions outside the Arctic. While there is general support for large contributions from both Eurasia and North America (Xie et al., 1999; Sharma et al., 2006), it is crucial to quantify the relative importance of emissions from various source regions in determining local pollutant levels. This will enable us to better understand the influence of past emission changes, such as the apparent maximum in North American BC emissions in the early 20th century (McConnell et al., 2007), and future changes such as the expected continuing decrease in emissions from mid/high latitude developed nations while emissions from lower latitude developing nations increase. Additionally, it will help to inform potential strategies to mitigate Arctic warming via short-lived pollutants (Quinn et al., 2007).

In this paper, we examine model simulations performed within the Task Force on Hemispheric Transport of Air Pollution (HTAP), a project to develop a fuller understanding of long-range transport of air pollution in support of the 51-nation Convention on

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Long-Range Transboundary Air Pollution. Using these simulations, we can analyze transport of a variety of idealized and actual pollutants to the Arctic in a large suite of models, allowing us to characterize the relative importance of emissions from different source regions as well as uncertainties in current understanding. As it is difficult to determine the source regions for Arctic pollutants directly from observations, and there have been some apparent inconsistencies in previous modeling studies (Law and Stohl, 2007), we believe that examining results from a large suite of models is a useful endeavor.

## 2 Description of simulations and analyses

A series of simulations were designed to explore source-receptor relationships (i.e. the contribution of emissions from one region, the source, to concentrations or deposition in a receptor region). The source regions were chosen to encompass the bulk of Northern Hemisphere emissions: Europe (EU: 10W-50 E, 25N-65N, which also includes North Africa), North America (NA: 125W-60W, 15N-55N), East Asia (EA: 95E-160 E, 15N-50N) and South Asia (SA: 50E-95E, 5N-35N) (Fig. 1). Northern Asia (Russia) was not included as a source region as its total emissions of most pollutants are comparatively small. However, given their proximity to the Arctic, emissions from this area can contribute substantially to Arctic pollution and so we caution that our analyses are not exhaustive. We define the Arctic poleward of 68 N as our receptor region. A base case simulation was initially performed using each model's own present-day emissions. Additional simulations then explored the response to a 20% reduction of anthropogenic emissions of nitrogen oxides ( $\text{NO}_x$ ) alone, carbon monoxide (CO) alone, or all anthropogenic ozone and aerosol precursors except methane from each of the four source regions. Participating models are listed in Table 1. We analyze the response of Arctic concentrations of trace gases and aerosols and deposition of BC in both Greenland and elsewhere in the Arctic.

As models used different base case anthropogenic emissions, the 20% perturbation

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tions differed in absolute amounts. Hence we generally analyze changes in Arctic abundances normalized by the regional emissions change between the control and the perturbation using direct emissions (CO, BC) or the dominant precursor (sulfur dioxide (SO<sub>2</sub>) for sulfate (SO<sub>4</sub>), NO<sub>x</sub> for ozone (O<sub>3</sub>)). Hereafter we refer to this quantity, in mixing ratio per Tg emission per season or year, as the Arctic sensitivity to source region emissions. With the exception of non-linearities in the response, this separates out the effect of intermodel differences in emissions. Uncertainties in emissions are of a different character than the physical uncertainties that we also explore, as the former depend on the inventories used to drive models while the latter are intrinsic to the models themselves.

The response to emissions changes in all four HTAP source regions were analyzed. All these simulations included a minimum of 6 months integration prior to analysis to allow for stabilization, followed by a year of integration with 2001 meteorology. Differences in meteorology were present, however, as models were driven by data from several reanalysis centers, or in some cases meteorology was internally-generated based on prescribed 2001 ocean surface conditions. Additionally, some models directly prescribed meteorology while others used linear relaxation towards meteorological fields. Note that the North Atlantic Oscillation index was weakly negative during 2001, while the broader Arctic Oscillation index showed a stronger negative value during winter, with weak positive values for most of the remainder of the year. These indices are reflective of the strength of the Northern Hemisphere westerly winds, with weaker winds associated with reduced transport to the Arctic (Eckhardt et al., 2003; Duncan and Bey, 2004; Sharma et al., 2006).

Idealized tracer simulations were also performed to isolate the effects of intermodel differences in transport from other factors affecting trace species distributions. For these simulations, all models used identical emissions of a CO-like tracer with a prescribed globally uniform lifetime of 25 days (though this is only about half the annual average global mean lifetime of CO). The range of model results in these simulations thus reflects only the variation in the transport algorithms used and in the meteorology

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used to drive the transport (which differed among models as discussed above). Emissions from different source regions were tagged. We examine the Arctic concentration of the regionally tagged tracer divided by the source region emission, analogous to the Arctic sensitivity described above (though these are absolute concentrations in a single run rather than a difference between a control and a perturbation run). These simulations did not include any spin-up, leading to substantial trends in total CO over the first several months.

All results for the Arctic are based on area-weighted averages. Results for Greenland are averaged over the entire Greenland land area, including the area south of that defined here as Arctic, neglecting model grid boxes near the coast that contain more ocean than land area. Surface values are those in the lowest model layer, while 500 and 250 hPa levels are interpolated from model output. All seasons refer to their boreal timing.

### 3 Modeled sensitivities, concentrations and deposition

In this section, we first consider the idealized simulations using a passive tracer with a prescribed lifetime (Sect. 3.1). We then analyze the results from the simulations using realistic chemistry and physics for both gases and aerosols (Sect. 3.2). Finally, we investigate model results for the deposition of black carbon to the Arctic (Sect. 3.3).

#### 3.1 Prescribed lifetime tracer

Transport of European emissions to the Arctic surface is clearly largest in winter (Fig. 2) based on results from 10 models (Table 1). This is despite the fact that these simulations did not include any spinup, and hence the values for the first several months of the runs are biased substantially low. Second year continuation runs performed by three of the ten models found that the winter Arctic sensitivity is biased low by up to a factor of 2 (though only about 10–25% for EU at the surface). This bias affected all



models, however, so should not greatly influence the model intercomparison.

During all seasons, the Arctic surface level is clearly most sensitive to European emissions (Fig. 2). In the middle troposphere, the sensitivities to emissions from Europe and North America are usually comparable, sensitivities to East Asian emissions are somewhat less, and sensitivities to South Asian emissions are quite small (probably because of the greater distance to the Arctic from this region (Fig. 1)). These results are consistent with the 'polar dome' or 'polar front' that impedes low-level transport from relatively warm and humid areas such as North America and East Asia into the Arctic during the cold months while allowing such transport at higher altitudes from those regions and at low-levels from Eurasia, which often lies within the polar dome (Law and Stohl, 2007; Klonecki et al., 2003). During summer, when the polar front is at its furthest north, emissions from East Asia, Europe and North America have a comparable influence on the Arctic surface (per unit emission), with a slightly larger contribution from Europe.

In the upper troposphere, the models tend to show comparable sensitivities for East Asian, European and North American emissions (and to a lesser extent SA), but usually with a larger spread of results than at other levels. Sensitivities in the upper troposphere are greatest in summer, consistent with the surface for Asian and North American emissions but opposite to the surface seasonality seen for European emissions.

### 3.2 Active gas and aerosol species

We now investigate the more realistic, but more complex, full gas and aerosol chemistry simulations. We sample only the models that performed the perturbation runs for a particular species (Table 1). The divergence in model results in the control run is extremely large in the Arctic. For example, CO varies by roughly a factor of 2–3 at all levels examined. Arctic sulfate varies across models by factors of 6 at the surface, 200 in the mid-troposphere, and 1000 in the upper troposphere. Though some models clearly must have quite unrealistic simulations, we purposefully do not exclude any models at this stage as our analysis attempts to identify the sources of this enormous

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divergence among models.

We first examine the total contribution from each source region to the annual average gas or aerosol amount in the Arctic. This includes the influence of variations in emission inventories among the models (Table 2). These variations are quite large, often as great as a factor of two between minimum and maximum. Surprisingly, the range of SO<sub>2</sub> and BC emissions used in the models is especially large for Europe compared to other regions. For the multimodel mean, we find that at the surface, European emissions dominate the Arctic abundance of sulfate and BC, and to a lesser extent CO (Table 3 and Fig. 3). Arctic surface ozone responds most strongly to NO<sub>x</sub> emissions from North America, with substantial responses to emissions from Europe and East Asia as well. In the mid-troposphere, sulfate abundances are again dominated by European emissions, but the contribution from East Asia is almost as large as that from Europe for BC. By the upper troposphere, both total sulfate and BC show the largest impact from East Asian emissions, especially for BC (Fig. 3). The amount of CO from each region also undergoes a shift with altitude, as European emissions become steadily less important relative to East Asian and North American emissions. The relative importance of regional NO<sub>x</sub> emission changes to Arctic ozone is less dependent upon altitude, with the largest contribution from North America at all levels.

We next turn to Arctic sensitivities (Arctic concentration change per unit source region emission change, hence removing the influence of emission inventory variations across models) rather than total Arctic concentrations, first examining seasonal sensitivities for CO, SO<sub>4</sub>, and BC (Fig. 4). The median Arctic surface sensitivities for all three species are greatest for European emissions, by roughly a factor of ~3–6 compared with the sensitivities to emissions from other regions. Median sensitivities in the mid-troposphere are again largest for European emissions in nearly all cases, by a few percent to a factor of two. In the upper troposphere, median sensitivities are comparable for East Asian, European and North American emissions. In many cases, sensitivities to South Asian emissions are substantially less than those for other regions. This is not the case for aerosols during winter and spring though, when sensitivities

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to South Asian emissions are large and sometimes greater than those for any other region. However, the range of sensitivities among models is quite large, especially for the aerosols.

Examining the CO sensitivity to the emissions with the greatest impact (EU, NA and EA), the range of annual average values is roughly a factor of 2–3 among the 11 models. The standard deviation is much smaller (~20–30%), indicating that most models are relatively consistent. The seasonality of the Arctic CO sensitivity depends on the source region. At the surface, the multimodel mean sensitivity to European emissions clearly maximizes in winter, while for North American and especially East Asian emissions the maximum sensitivity is in spring. These two regions also differ in their seasonality, however, with the minimum sensitivity in fall for East Asian emissions but in summer for North American emissions.

For surface sulfate, Arctic sensitivities in individual models vary greatly. For the annual average, the range spans 2.8 to 17.4 pptm/(Tg S)/season. (Note that the annual average values are in units of pptm/Tg/season for comparison with seasonal sensitivities. Values in pptm/Tg/year are 1/4 of these season numbers). Interestingly, the models separate into two groups: of the 14 models, 6 have annual average sensitivities below 4 pptm/(Tg S)/season, while the other 8 have sensitivities of 7.2–17.4 pptm/(Tg S)/season. Seasonal surface sensitivities show an even larger spread (Fig. 4). Median sensitivities to European emissions are comparable in all seasons though the spread in the central 50% of models is greatest in winter and spring. Sulfate sensitivities to East Asian and North American emissions maximize in spring, as for CO. In the mid-troposphere, sensitivities are generally largest for European emissions, while in the upper troposphere they are greatest for South Asian emissions in spring and East Asian emissions during other seasons.

BC sensitivities show spring maxima for surface responses to East Asian and North American emissions, as for sulfate and CO. For European emissions, however, BC sensitivity shows a strong winter maximum and a fall sensitivity that is also enhanced over the spring and summer values. The mean winter sensitivity to European emissions

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of 26 ppbm/(Tg C)/season is much larger than the sensitivities to European emissions during the other seasons (8–15 ppbm/(Tg C)/season mean). The enhanced winter sensitivity results from both faster transport during winter and slower removal at this time as the Arctic is stable and dry. During spring, summer and fall, the mid-troposphere, like the surface, is most sensitive to European BC emissions. During winter, however, sensitivity to North American and European emissions is almost identical. Interestingly, the seasonality of sensitivity can vary with altitude: the sensitivity of surface BC to European emissions is greatest in winter, while the sensitivity of mid-tropospheric BC to European emissions maximizes in summer (sulfate sensitivities show fairly similar behavior). North American emissions are also most effective at influencing the Arctic mid-troposphere during summer, but the sensitivity to East Asian emissions maximizes in spring. Sensitivities in the upper troposphere maximize in summer for EA, EU and NA. Though the multimodel mean surface and mid-tropospheric sensitivities are clearly greatest for European emissions, the annual average BC sensitivity to European emissions varies greatly among models: from 0.6 to 7.9 ppbm/(Tg C)/year for the surface and from 0.1 to 12.6 ppbm/(Tg C)/year at 500 hPa.

The sensitivity of Arctic surface ozone to source region  $\text{NO}_x$  emissions is quite different than that for the other species examined here (Fig. 4). Note that MOZECH and UM-CAM were excluded from the  $\text{O}_3/\text{NO}_x$  analysis due to imbalances in their nitrogen budget diagnostics. Sensitivity to winter European emissions is negative for most models (i.e. reduced  $\text{NO}_x$  emissions leads to more Arctic ozone). This results from direct reaction of  $\text{NO}_x$  with ozone in the relatively dark conditions that much of the high-latitude European emissions encounter. The cancellation of negative winter and positive non-winter ozone sensitivities leads to a lower annual average European influence on Arctic surface ozone than for other species (Table 3). Spring surface ozone concentrations show comparable sensitivities to East Asian, European and North American emissions, while summer concentrations are most sensitive to European and fall to European and North American emissions. During winter, sensitivities to South Asian emissions are nearly as large as those for East Asian and North American emissions. Sensitivities in

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the mid-troposphere show a similar pattern to those seen at the surface (Fig. 4). The upper troposphere shows comparable sensitivities for all four of the source regions, with greatest sensitivity to South Asian emissions in winter and spring, though North American emissions have the largest annual average influence (Fig. 3).

5 The magnitude of the sensitivity increases with altitude for  $\text{SO}_4$  and  $\text{O}_3$ , stays roughly constant for BC, and decreases for CO. This may reflect the greater removal of soluble aerosols and ozone precursors at low levels relative to insoluble CO. In addition, both  $\text{SO}_4$  and  $\text{O}_3$  are produced photochemically at higher altitudes, while CO is photochemically removed aloft.

10 A critical result of the analysis is that for both sensitivities and totals, discrepancies between models are systematic, so that the relative importance of different regions is robust despite the large differences among models in the magnitude of the contribution from a particular region. For example, every participating model finds the largest total contribution to annual average surface  $\text{SO}_4$ , BC, and CO to be from Europe. Similarly,  
15 all models find that in the upper troposphere, East Asia is the largest annually-averaged source for BC, with all but 3 giving first rank to East Asia for sulfate and CO as well. Looking at annual totals, 9 out of 11 models have a larger contribution to 500 hPa Arctic ozone from North America than from Europe even though the standard deviations of these overlap substantially (Table 3). For sensitivities, every model finds that the Arctic  
20 sensitivity during winter, fall and spring for surface  $\text{SO}_4$ , BC and CO is largest for European emissions. This holds even during summer for sulfate and BC, while for CO all but 1 model has the greatest sensitivity to European emissions.

### 3.3 Arctic deposition of black carbon

In addition to atmospheric concentrations, deposition of BC to the Arctic is of particular  
25 interest due to its climate impact, as discussed previously. We now explore the relative importance of the various source regions to BC deposition to Greenland and to the rest of the Arctic, and use the multimodel results to characterize the robustness of these results. We examine both the total BC deposited and also the BC deposition

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sensitivity by calculating the Tg deposited per unit area per Tg source region emission. Deposition is calculated on all surfaces, including open ocean, though albedo will be affected by the flux to snow and ice surfaces.

Deposition of BC to the Arctic (excluding Greenland) is most sensitive to emissions from Europe in every season (Fig. 5), and is generally quite similar to the BC surface mixing ratio sensitivity (Sect. 3.2). That sensitivity to European emissions is greatest is clear even though the spread in model sensitivities is very large, more than an order of magnitude in most seasons for Europe, for example. The large range often results from just one or two models. For example, the deposition sensitivities for the Arctic (except Greenland) during summer for European emissions are within  $0.09$  to  $0.32 \text{ m}^{-2} 10^{-11}$  in 7 models, while the remaining two have values of  $0.76$  and  $2.14 \text{ m}^{-2} 10^{-11}$  (ECHAM5-HAMMOZ is excluded from the BC deposition analyses owing to apparent problems in their deposition diagnostics). However, even the range within the central 50% of models is substantial in other seasons (Fig. 5). Sensitivities are largest in fall and winter for European emissions, but in spring, summer and fall for North American emissions and in spring for East Asian emissions. Examining the BC deposited per unit BC emitted (i.e. multiplying the values in Fig. 5 by area), the multi-model mean percentage of emissions that are deposited in the entire Arctic (including Greenland) is 0.1% for South Asian, 0.5% for East Asian, 1.0% for North American and 3.6% for European emissions. The deposition of BC is much more sensitive to the location of emissions than is the burden of BC in the Arctic. The multi-model mean percentage of emissions that reside in the annual average Arctic BC burden varies by only a factor of 4 across regions: 0.02% SA, 0.03% EA, 0.04% NA and 0.07% EU (regional emissions contributions to the Arctic burden of sulfate are 0.02% SA, 0.03% EA, 0.03% NA and 0.05% EU, quite similar to BC, while for longer-lived CO they are 0.6% SA, 1.2% EA, 1.6% NA and 2.1% EU).

Deposition of BC to Greenland shows different sensitivities compared with the rest of the Arctic, with the largest response to North American emissions in non-winter seasons. This results from the high topography of Greenland, which allows the inflow

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of air from the relatively warm and moist North American, and to a lesser extent East Asian, source areas to occur more easily there than in the rest of the Arctic (Stohl, 2006).

Examining the total BC deposition response to 20% regional emissions changes, we find similar results for the Arctic excluding Greenland as for the sensitivity (compare Figs. 6 and 5). Most significantly, total deposition is greatest from Europe in every season. Again the spread of results is large, but the central 50% of models are distinctly separated for Europe from other regions in all seasons but spring. In that season, East Asian emissions take on greater importance as they are large and the sensitivity to East Asian emissions maximizes in spring (Fig. 5). For the annual average, total deposition to the Arctic outside of Greenland is clearly dominated by European emissions (Table 4). We reiterate that emissions from Northern Asia (Russia) were not studied in these analyses.

The change in total deposition to Greenland in response to 20% regional emissions changes is roughly evenly split between the impact of European and North American emissions (Fig. 6). Deposition of BC from East Asia is as large or nearly as large as that from these regions in spring and summer, though not in other seasons or in the annual average. The spread of results for total deposition to Greenland is substantially larger for totals than for sensitivities for East Asia and Europe, reflecting the large variation between models in emissions from or within these regions. Looking at the annual average total deposition to Greenland, it appears at first that the uncertainties are too large to allow determination of the relative importance of emissions from the various source regions (Table 4). However, the distribution of results is neither normally distributed nor random. To test the impact of “outlier” models, we calculated deposition statistics leaving out the models with the lowest and highest deposition rates (Table 4). The model with the largest deposition contributes a large fraction of the standard deviation. The lowest does not, however, as the distribution of results is highly skewed towards values considerably above the mean. To test for systematic effects across regions, we also examined the relative importance of each region across models. While

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one standard deviation of the deposition values for a given region nearly always encompasses the values for the other regions, in fact the model-to-model differences are systematic. Every one of the 10 models has the identical order: greatest deposition to Greenland from North American emissions, followed by European, East Asian and lastly South Asian emissions. Deposition to the Arctic (exclusive of Greenland) is similarly skewed (Table 4) and robust in the regional rankings across models. Hence as for atmospheric mixing ratios and concentration sensitivities (Sect. 3.2), the relative contribution of emissions from the various source regions to BC deposition can be determined with much higher confidence than the magnitude for any particular region.

## 4 Comparison with Arctic observations

Observational datasets are quite limited in the Arctic, making it challenging to reliably evaluate models in this region. Nevertheless, it is worth investigating how well the models perform and how our results are influenced by any models which appear to be clearly unrealistic in their Arctic simulations. In this section, we compare the modeled and measured seasonal cycles of surface CO, ozone, sulfate and BC for selected stations in the Arctic. Root-mean-square (RMS) errors between the monthly mean modeled and observed values are used to evaluate the models, though this is clearly only one possible measure of model/observation agreement. We then evaluate the influence of screening out less realistic models in Sect. 5.

The models exhibit a large spread for CO at Barrow and Alert (Fig. 7), though most have a fairly reasonable seasonal cycle based on comparisons with observations (Novelli et al., 1998). Many of the models do a good job of reproducing summer and fall CO amounts, but nearly all underestimate the late winter-early spring maximum (as in previous studies, e.g. Shindell et al., 2006). All models have average RMS errors for the two sites of between 17 and 40 ppbv, with the exception of two that have RMS values of 54 and 83 ppbv. We note that no model stands out as substantially better than the others (the model with the second lowest RMS error has a value only 3 ppbv

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greater than the lowest score). While the EMEP model stands out from the others with clearly larger values during fall (Fig. 7), it is not obviously better or worse in comparison with observations over the full annual cycle. Note also that none of the outlying models (those labeled in Fig. 7) has CO emissions distinctly different from the other models.

5 Hence excluding the two models with highest RMS error provides a reasonable subset for repeating the CO analyses.

Comparison of ozone observations (based on updates from Oltmans and Levy, 1994) with the models shows that the simulations again have a fairly wide spread. Modeled values are generally reasonable during summer and fall at Barrow, though some models have underestimates, but agreement is poor during winter and spring. Nearly all models underpredict ozone at Summit. Those that overestimate ozone at Barrow during spring often do a better job at Summit, as they fail to capture the large observed springtime contrast between these two sites. This leads to comparable error scores to other models. All models have an RMS error of 7–12 ppbv except for STOCHEM-HadGEM1 with a value of 21 ppbv. We find that exclusion of a single model, however, does not appreciable change the results presented previously.

Sulfate observations present several challenges. Among these are that sulfate totals are observed, and the sea-salt component has to be estimated and removed to obtain non-sea salt sulfate concentrations. We use data from the EMEP network's station on Spitsbergen (Hjellbrekke and Fjæraa, 2007). Other Arctic data covering appropriate years was difficult to find, so we also compare with measurements taken at Denali National Park (from the Interagency Monitoring of Protected Visual Environments: IMPROVE, <http://vista.cira.colostate.edu/improve>), although at 64° N this is just outside the Arctic region we use here. The models generally perform poorly in simulating Arctic sulfate. Most substantially underestimate Arctic concentrations, with more than an order of magnitude underestimation in several models. Many that show sulfate concentrations of about the right magnitude have seasonal cycles that peak in summer or fall, while the observations show a spring maximum. None of the models produce a spring maximum of the right magnitude at Denali, though several do a better job in capturing

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the spring peak at Spitsbergen (Fig. 7). This leads to RMS error values that are fairly large for all models, with multi-model means of 249 (Denali) and 272 (Spitsbergen) pptm. However, models cluster in two groups, with none having average values for the two sites between 200 and 240. Hence we can test if the subset of models with RMS values below 200 pptm yields a different result than the full suite of models.

We also attempted to evaluate the simulation of BC in the models. Comparison with observations from Sharma et al. (2006) suggests that models greatly underpredict BC in the Arctic (Fig. 7). However, the available measurements are in fact equivalent BC (EBC), which is obtained by converting light absorbed by particles accumulated on a filter in a ground-based instrument to BC concentrations. In addition to BC, other light absorbing species such as OC and dust influence the measurements, so the EBC would tend to be high relative to actual BC. This is consistent with the sign of the model/observations difference (Fig. 7), though a substantial underestimate in the models remains possible. Models also appear to substantially underestimate BC in comparison with IMPROVE data from Barrow (updated from Bodhaine, 1995), which itself differs significantly from the Sharma et al. (2006) data. The Barrow data is also derived from optical absorption measurements. As it is not known precisely how the actual BC (or OC or dust) mixing ratios relate to the observed optical properties, it is extremely difficult to quantitatively evaluate the models. Given the large apparent discrepancies for BC for all models, we conclude that it is not feasible to determine the relative realism of the models using currently available data.

## 5 Causes of intermodel variations

In this section, we investigate the role of model-to-model differences in transport, photochemistry and deposition, and emissions in creating the diversity of results seen in the Arctic. This is accomplished by comparison of the prescribed lifetime simulations with the full chemistry and physics simulations and by examining the correlation between Arctic concentrations and diagnostics such as residence times.

## 5.1 Isolating processes governing variations in CO and ozone

We first compare the intermodel variability in Arctic sensitivities in the run with the prescribed lifetime “CO-like” tracer to that with realistic CO. Note that the different lifetimes of CO in the prescribed lifetime and full chemistry runs may influence the variations in transport among models. As intermodel variations in transport would be unimportant for a very long-lived, well-mixed tracer, we expect that the importance of intermodel variations in transport is inversely related to lifetime, and hence the values seen in the prescribed 25-day lifetime tracer would be greater than the true variations for CO and less than those for shorter-lived species. Future prescribed lifetime experiments planned within the HTAP project will characterize the interaction between lifetime and transport uncertainties. As the two sets of experiments do not use directly comparable CO, we analyze the standard deviation as a percentage of the mean response (the fractional variation) among models. The analysis is restricted to the subset of 9 models that performed both the prescribed lifetime and full chemistry runs.

The fractional variation of sensitivity is nearly always larger in the full chemistry analyses than in the prescribed lifetime case, with the lone exceptions of surface sensitivity to South Asian emissions (numerical values in Fig. 8). In that case, intermodel differences in chemistry may offset some of the difference in modeled transport, or, as noted, the prescribed lifetime results may be overestimates of the intermodel variations for actual CO. Examining the sensitivity to emissions from other regions, the relative size of the fractional variations in the prescribed lifetime and full chemistry runs depends on the altitude analyzed and the source region. At the Arctic surface, the intermodel fractional variation in the prescribed lifetime runs is 12–14%, more than half that in the full chemistry runs (16–24%) for all three other regions (Fig. 8). This indicates that differences in modeled transport to the Arctic play an important role in CO near the surface. In the middle and upper troposphere, transport and chemical oxidation by OH contribute a comparable amount to intermodel differences in Arctic CO. At the surface and in the mid-troposphere, adding in the intermodel variation in emissions

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(i.e. no longer normalizing by emissions) leads to larger fractional variances across the models. This is especially so for East Asia, where including the intermodel variation in emissions nearly triples the fractional variance of the Arctic response at the surface and middle troposphere across models. The effects are smaller for emissions from Europe or North America, where emissions variations add ~5–13% to the fractional variance, a comparable range to that from transport (8–14%) and oxidation (1–11%) variations among models. Emissions uncertainties from South Asia have an even smaller impact than those from Europe or North America, barely changing the fractional variance.

Thus the intermodel variation in the influence of source region CO emissions on the Arctic surface is dominated by emissions for East Asia, by transport for South Asia, and all three terms (transport, oxidation and emissions) play comparable roles for Europe and North America. In the middle altitudes, East Asian intermodel variations are dominated by emissions, transport plays a major role in North American and South Asian variations, and European variations are influenced by all three. In the upper troposphere, transport and oxidation differences dominate the intermodel fractional variations. Thus the main change with height is the gradually increasing importance of intermodel differences in oxidation at higher levels. We note, however, that while the 250 hPa intermodel differences are important, the variation across the central 50% of models in CO sensitivity in the upper troposphere is only ~40%, among the smallest range for any species at any level (Fig. 4).

We now investigate the dependence of the results on the quality of the model's Arctic CO simulation. Including all models, the fractional variation is 20–26% for surface and mid-troposphere sensitivities to European, East Asian and North American emissions, and is 31–35% for SA emissions. Using only the subset of models showing better agreement with observations (9 of 11 models), it decreases to 11–13% for surface and mid-troposphere sensitivities to East Asian and North American CO emissions, 16% and 22% for surface and mid-troposphere sensitivities to European emission, respectively, and 23–25% for SA emissions. There is no appreciable difference between using all models or restricting the analysis to the subset when calculating the

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standard deviations in the upper troposphere. In either case these are 24–29% for European, East Asian and North American emissions. The sensitivity to South Asian CO emissions in the upper troposphere shows a very large standard deviation across models (45–47%), perhaps related to variations in model simulations of tropical convective transport. However, its contribution per unit emission is relatively small. Hence screening models by their ability to match observations can substantially reduce the intermodel variations even though in this case only 2 models were removed. We conclude that the Arctic-wide response to source region emissions perturbations can be simulated relatively reliably for a long-lived species such as CO. This is especially true for quality-screened models, in which case fractional variations in the mid and lower troposphere are 22% or less for CO sensitivity to emissions from Europe, and 13% or less for emissions from East Asia or North America.

Though model results are relatively consistent for CO, it is interesting to examine the relationship across models between the Arctic sensitivities and the global mean chemical lifetime (lifetimes for portions of the globe could not be calculated using the available diagnostics). A high correlation would indicate that the removal rates of CO (by oxidation) play an important role in intermodel variations in sensitivity. We find that either using all models or the quality-screened subset there is little correlation between CO sensitivity and global mean lifetime for the surface and mid-troposphere, except for South Asian emissions (Table 5). There is some correlation for all regions in the upper troposphere ( $R^2$  0.4–0.7). Hence it appears that the CO chemical lifetime, a measure of the CO oxidation rate, does not play a large role in determining the sensitivity of the Arctic to NA, EU and EA emissions perturbations below the upper troposphere. These results are consistent with the increasing importance with height of oxidation seen in the comparison between the prescribed lifetime and full chemistry simulations. For emissions from South Asia, which have to travel further to the Arctic, the chemical lifetime does appear to play an important role, especially in the mid and upper troposphere.

Ozone's response to  $\text{NO}_x$  emissions perturbations can be of either sign, indicating

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non-linearities in chemistry that preclude explanation via linear correlation analysis of its lifetime. We can, however examine correlations between Arctic ozone sensitivity and intermodel variations in ozone dry deposition or transport (using the passive tracer simulations for the latter). We find that model-to-model variations in dry deposition account for little of the spread in Arctic ozone sensitivity to  $\text{NO}_x$  perturbations, even at the surface ( $R^2 < 0.3$  at all levels,  $< 0.1$  at surface). Correlations between Arctic ozone sensitivity and transport are similarly weak at the surface and 250 hPa ( $R^2 < 0.25$ ). In the mid-troposphere (500 hPa), however, correlations are  $R^2 = 0.4$  to  $0.5$  for NA, EA and SA, indicating that at those levels transport variations account for roughly half the intermodel variation in Arctic ozone sensitivity. At other levels, however, it appears that model-to-model differences in the non-linear ozone photochemistry must play the dominant role in the intermodel spread of sensitivities. Consistent with this, annual mean ozone sensitivity to  $\text{NO}_x$  perturbations shows a variation across models of 35–80% of the mean, which is substantially larger than the 8–31% in the prescribed lifetime tracer experiments

## 5.2 Isolating processes governing variations in aerosols

We examine the relationship between the Arctic concentrations and the aerosol lifetimes, as for  $\text{CO}$ . For aerosols, however, we are able to calculate the global residence time for regional emissions perturbations. These are determined from the change in burden over the change in removal rates in the regional perturbation experiments. Note that this calculation implicitly assumes that the residence time is the same in the two experiments, which given the relatively small emissions perturbations imposed in our experiments should be a good approximation. We also compare with the prescribed lifetime tracer results to estimate the relative importance of transport variations among models to the total range in results. We note that the lifetime for the prescribed tracer (25 days) is several times longer than that for aerosols, and hence, as discussed in Sect. 5.1, the intermodel transport variations for aerosols are likely larger than those in the prescribed lifetime simulation, and perhaps substantially so. Additionally, the sol-

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ability of some aerosols links them to the hydrologic cycle much more closely than for CO, so the comparison with the prescribed lifetime tracer results isolates the influence of dry transport (i.e. excluding transport of species in the aqueous phase).

The range of intermodel variations in Arctic sensitivity is much larger for BC than for CO (Fig. 4). The intermodel variation in residence time for BC among models is roughly a factor of 2 and accounts for most of the spread (Table 5). This variation is much greater than the variation in efficiency of dry transport to the Arctic at most levels from any region as seen in the prescribed lifetime simulations (e.g. Fig. 8). Hence the other factors affecting residence time, including aerosol aging from hydrophobic to hydrophilic and rainout/washout of the aerosols, appear to play important roles in governing the Arctic sensitivity to regional BC emissions from middle to higher Northern latitudes (EU, NA, and EA). In other words, the large variations in how long BC remains in the global atmosphere seems to be more important in determining how much reaches the Arctic than are transport differences or local Arctic removal processes (which contribute only a minor fraction of the global removal). This result is consistent with the strong sensitivity in the export efficiency of Asian BC to the conversion lifetime from hydrophobic to hydrophilic seen in a study based on 2001 aircraft data (Park et al., 2005). For emissions from South Asia, the global annual mean residence time of BC is less closely correlated with the Arctic sensitivity. Hence for emissions from this region, both intermodel variations in BC residence times and in transport appear to be important factors in creating model diversity.

For sulfate, the fractional variation in annual average sensitivity across models is ~68–112%, much greater than that seen in the prescribed lifetime runs where fractional variations were only ~8–31%. This suggests that for sulfate, as for BC, variations in large-scale aerosol physics (removal of sulfate and/or SO<sub>2</sub>, oxidation of SO<sub>2</sub>, etc.) account for a major portion of the divergence between models. When intermodel emissions variations are not removed from the sulfate response calculations, the annual average fractional variance increases only modestly (~10%), to 79–120% across the models. Thus emissions differences appear to play a minor role in the model-to-model

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variations.

Sulfate's global mean residence time in the models ranges from 2.7 to 11.2 days. Aside from the two models with these values, the other 12 all have lifetimes between 3.2 and 7.4 days. Hence the spread in global residence times is small compared with the spread in Arctic sensitivities (Fig. 4). Correlations between these two quantities are fairly weak at the surface, but more significant aloft when examining all models (Table 5). Using only those in the subset with lower RMS error scores against observations (which screens out the models with 2.7 and 11.2 day lifetimes), the correlations increase at the surface in some cases, but decrease at 500 and 250 hPa. Even with the quality-screened subset of models, the residence time for regional emissions perturbations typically accounts for at most 20–50% of the variance in lower tropospheric Arctic sensitivities, and often 0–10%. Hence while variations in residence time account for a substantial fraction of the intermodel variance in Arctic sensitivities across all models, they can explain only a modest portion of the variance in the subset. In the latter models, the variation in residence time is relatively small, so that processes such as wet removal of sulfate or in-cloud oxidation farther from the source region may be important in controlling how much sulfate reaches the Arctic even though they may not greatly affect the global residence time for regional emissions perturbations. Transport variations between models may also be playing a larger role than indicated by the comparison with the longer-lived prescribed lifetime tracer. Note that residence times are somewhat longer for European emissions, consistent with their larger impact on the Arctic. We also point out that sulfate changes are a function of both aerosol and oxidant precursor changes in these experiments, as precursors to both were changed simultaneously in the HTAP runs. This may at least partially explain why sulfate residence times are not as well correlated with Arctic sensitivities as are BC's. Hence diagnosing the physical processes responsible for the large spread in sulfate sensitivities will require much deeper investigation into model processes using additional diagnostics not available in the HTAP archive, and would benefit from additional simulations perturbing only aerosol precursors.

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Overall, the comparison between the prescribed lifetime tracer and full chemistry simulations and the analyses of the correlation between residence times and Arctic concentrations both support the conclusion that dry transport differences among the models play a major role in the intermodel variations of insoluble, relatively long-lived CO. They are similarly important contributors to the model-to-model differences in mid-tropospheric ozone. However, these appear to be less important contributors to the intermodel variations in the Arctic sensitivity to aerosol emissions, for which uncertainties in aerosol physics and wet removal play important roles.

We also examined the relationship between horizontal resolution in the models and their representations of transport and of trace species in general. Horizontal resolution, using latitude, ranges from 1 to 5 degrees (for the BC lifetime analysis, it ranges only from 1 to 2.8 degrees). We find  $R^2$  correlations with resolution (using latitude) to be extremely low for lifetimes and sensitivities. Hence there is no straightforward correlation with resolution.

## 6 Discussion and conclusions

The spread in model results for Arctic pollutants is very large for both gaseous species and aerosols. Differences in modeled transport, chemistry and emissions all contribute to this spread, which makes climate and composition projections for the Arctic extremely challenging.

This study has identified the largest contributing factors to the diversity of model results. We have shown that for sulfate and BC (including deposition of the latter), uncertainties in modeling of aerosol physics are extremely important, with lesser roles for emissions and probably for transport. Further studies to determine precisely which physical processes play the largest role, such as those suggested by (Textor et al., 2006), would help prioritize research. In contrast, for CO, transport and emissions are important drivers of uncertainty in simulating surface responses to source region emissions, while transport, emissions and oxidation rates all play comparable roles at

higher altitudes. For ozone, our analysis suggests that transport plays a substantial role in the intermodel variations in sensitivity, but that photochemical differences among the models appears to be the dominant contributor.

Our results for aerosols are consistent with earlier intercomparisons. These showed that the diversities in aerosol mass depend largely on differences in transport and the parameterizations of internal aerosol processes, and only to a lesser extent on their (precursor) emissions (Textor et al., 2007). These results held true for both the global aerosol load and the polar ( $>80^\circ$  in both hemispheres) fraction. Our results also suggest that the contribution of intermodel transport differences to disparities in Arctic aerosol loading is relatively small, reinforcing the conclusion that aerosol and cloud physics (e.g. removal, chemistry and microphysics) is the principle source of uncertainty in modeling the distributions of these species in the Arctic.

For cases in which transport plays a substantial role in intermodel variability, such as CO or mid-tropospheric ozone, intercomparison among different models driven by the same meteorological fields would help determine the underlying reason for the range of results (complimenting studies of a single model driven by multiple meteorological fields, such as Liu et al., 2007). Differences in convection certainly contribute to transport variations among models. Model numerical schemes could also play a role, though algorithms such as conservation of second-order moments have been shown to generally transport trace species quite well, preserving gradients and not being too diffusive (Prather, 1986). However, this merits further study as many models may use less capable transport schemes. Additionally, the degree of agreement between chemical-transport models driven by offline meteorological fields and general circulation models that are relaxed towards offline meteorological fields remains to be characterized. Our comparison shows no clear effect of horizontal resolution.

Although the intermodel variations in transport to the Arctic are large, many of them are systematic across models so that differences between sensitivities to emissions from various regions are robust across models. In particular, we find that Arctic surface concentrations of BC, sulfate, and CO are substantially more sensitive to European

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emissions than to those from other regions. Similar results are obtained for the mid-troposphere (500 hPa), though the difference in sensitivities between Europe and other regions is not as large as for the surface. Hence per unit Tg emission change, European emissions are the most important. We expect that Arctic sensitivities to emissions from Northern Asia would be generally similar to their European counterparts given the similarity in proximity and meteorological conditions.

Arctic surface concentrations are most sensitive to European emissions during winter for CO, sulfate and BC. In the middle troposphere, sensitivity to European emissions is greatest in summer for aerosols. Concentrations are most sensitive to East Asian emissions during spring for BC, sulfate, and CO at both the surface and 500 hPa. Hence the relative importance of emissions from different regions varies seasonally. For surface ozone, Arctic concentrations are most sensitive to European emissions of NO<sub>x</sub> during summer, but sensitivities are comparable in fall for European and North American emissions, and in spring for East Asian, European and North American emissions. In the upper troposphere, concentrations for all species typically show comparable sensitivity to emissions from all four source regions, though there is a general tendency for a lower sensitivity to South Asian emissions (especially for CO).

The deposition of BC to the Arctic outside of Greenland is most sensitive to emissions from Europe in all seasons. In contrast, deposition of BC to Greenland is most sensitive to North American emissions, except during winter when sensitivity to European emissions becomes comparable. Total deposition of BC, rather than per unit emission, is again greater from Europe than the other regions for the Arctic exclusive of Greenland. Annual mean total BC deposition onto Greenland is greatest from North America and Europe, which are nearly equal, with a substantial but lesser contribution from East Asia. These conclusions are robust across the models examined here. Total springtime deposition to Greenland is primarily due to emissions from North America and East Asia, when Greenland is less affected by European emissions than in other seasons. As springtime deposition appears to be especially effective in inducing large snow-albedo feedbacks (Flanner et al., 2007), this suggests an enhanced role

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in Greenland climatic forcing for East Asian and North American emissions relative to their annual mean contribution to deposition.

The recent recovery of ice core records from Greenland containing BC (McConnell et al., 2007) may allow better estimates of historical BC emissions. The results presented here indicate that even without including Russian emissions, North America is responsible for less than half the BC deposition onto Greenland (Table 4). Hence the ice core record may indeed reflect very large emissions during the early 20th century from Eastern North America (McConnell et al., 2007), but it could also include the effects of historical emissions from other regions. Further analysis of historical emission trends could help clarify this issue by matching the onset and duration of the early 20th century BC deposition maximum seen in the ice core record.

Previous work has discussed the apparently conflicting results on transport of BC to the Arctic (Law and Stohl, 2007). The results of (Koch and Hansen, 2005) indicated that Arctic BC optical thickness results mostly from Asian emissions (excluding Russia, so roughly corresponding to our SA+EA). Impacts from European and North American emissions were roughly half to one-third of the Asian ones, and Asian emissions also played a major role in the low altitude springtime Arctic Haze. In contrast, (Stohl, 2006) found that transport from Europe was much more effective than from South and East Asia. The mean BC source region emissions in HTAP are: SA 0.91, EA 2.01, NA .71, EU 1.23 Tg yr<sup>-1</sup>. In (Koch and Hansen, 2005), they are: SA+EA 2.08, EU 0.47, NA 0.39 Tg yr<sup>-1</sup>. Using either set of emissions and the sensitivities found here, BC in the upper troposphere is indeed dominated by Asian emissions (as in Table 3). In the mid-troposphere, Asian emissions dominate during winter and spring, have comparable impact to European emissions in fall, and are less important in summer using HTAP emissions. Using those of (Koch and Hansen, 2005), Asian emissions would be most important in all seasons. Examining springtime low altitude BC pollution (contributing to Arctic Haze), European sources contribute ~70% more than Asian sources in the HTAP models. However, using the HTAP sensitivities and the Koch and Hansen (2005) emissions yields comparable impacts from Europe and Asia. Hence although the GISS

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model used by (Koch and Hansen, 2005) may transport BC to the Arctic more efficiently than other models (including the HTAP models simulating BC), their results for the relative importance of emissions from different regions are generally supported by the BC model simulations analyzed here, with differences largely arising from the differing emission inventories used. We find no contradiction between the large impacts of Asian BC aloft in the Arctic and the dominant role of European emissions on surface BC.

The results presented here help to characterize the relative importance of emissions from various source regions in seasonal and annual Arctic pollution. It remains an open question how these sensitivities may change in the future. As climate continues to change, Arctic temperatures are projected to increase faster than those at lower latitudes. This would reduce the temperature difference between the Arctic and pollution source regions, enhancing low-altitude transport of pollution into the Arctic. This might lead to increased pollutant concentrations and, if these were primarily climate warming agents, a further increase in surface temperatures. Additionally, large-scale circulation patterns are also projected to respond to climate change. These may respond to the projected increase in the temperature gradient in the vicinity of the tropopause rather than the decreased gradient near the surface (Shindell et al., 2001). The Arctic may be strongly affected by changes in the Northern Hemisphere westerlies associated with the North Atlantic Oscillation/Northern Annular Mode (Eckhardt et al., 2003; Duncan and Bey, 2004), which are projected to accelerate in the future (Miller et al., 2006). These westerlies have been shown to substantially enhance pollutant transport to the Arctic, at least from some regions (Sharma et al., 2006). Hence transport from highly polluted source regions may become more frequent in the future.

At the same time, emissions will also be changing. Projected increases from East Asia would be especially effective in causing more springtime ozone, sulfate, BC and CO both at the Arctic surface and in the mid-troposphere. Reductions in emissions from developed nations in North America and Europe would cause decreases in surface level CO and BC that would be especially pronounced in winter. This would also substantially reduce BC deposition onto Greenland, though the reduction might

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be largely offset, especially during spring and summer, by Asian emission increases. Surface sulfate would be reduced most strongly in winter and spring with decreased emissions from industrialized countries, while ozone concentrations would decrease most in non-winter seasons. Hence changes in the seasonal cycle of surface CO and BC, for example, with a reduced winter-to-spring gradient, could result from a shift in emissions from the developed to the developing world.

Understanding of future Arctic pollution levels will require simulations incorporating both changing climate and emissions. Our confidence in the results of such simulations could be greatly improved by resolving some of the apparent discrepancies between model results and observations, especially for aerosols. Better measurements of Arctic aerosols are clearly required. Hopefully the activities of the International Polar Year 2007–2009 will substantially increase our knowledge of the Arctic, and a heightened focus on the Arctic by the community will lead to at least some of these measurements being maintained over the long-term. Nevertheless, the current results are robust across models in many respects, allowing better understanding of how various types of pollutants arrive in the Arctic and influence climate and air quality.

*Acknowledgements.* We thank the NASA Atmospheric Chemistry Modeling and Analysis Program for support, and D. Henze and D. Koch for comments. MGS and KJP were funded by the UK Defra under contract AQ0409, and were also supported by the Joint Defra and MoD Programme, (Defra) GA01101 (MoD) CBC/2B/0417\_Annex C5. This work was performed under the umbrella of the Task Force – Hemispheric Transport of Air Pollution ([www.htap.org](http://www.htap.org)).

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**Table 1.** Models simulations used in the analyses.

Model	Gas-phase	Aerosols	Prescribed lifetime	Horizontal Resolution
1. CAMCHEM	NO <sub>x</sub> , CO	SO <sub>2</sub> , BC	Y	1.9
2. ECHAM5-HAMMOZ		SO <sub>2</sub> , BC		2.8
3. EMEP	NO <sub>x</sub> , CO	SO <sub>2</sub>		1.0
4. FRSGC/UCI	NO <sub>x</sub> , CO		Y	2.8
5. GEOSChem	NO <sub>x</sub>	SO <sub>2</sub> , BC		2.0
6. GISS-PUCCINI	NO <sub>x</sub> , CO	SO <sub>2</sub>	Y	4.0
7. GMI	NO <sub>x</sub> , CO	SO <sub>2</sub> , BC	Y	2.0
8. GOCART-2		SO <sub>2</sub> , BC		2.0
9. LMDz4-INCA		SO <sub>2</sub> , BC		2.5
10. LLNL-IMPACT	NO <sub>x</sub> , CO	SO <sub>2</sub> , BC		2.0
11. MOZARTGFDL	NO <sub>x</sub> , CO	SO <sub>2</sub> , BC	Y	1.9
12. MOZECH	NO <sub>x</sub> , CO		Y	2.8
13. SPRINTARS		SO <sub>2</sub> , BC		2.8
14. STOCHEM-HadGEM1	NO <sub>x</sub> , CO		Y	3.8
15. STOCHEM-HadAM3	NO <sub>x</sub> , CO	SO <sub>2</sub>	Y	5.0
16. TM5-JRC	NO <sub>x</sub>	SO <sub>2</sub> , BC	Y	1.0
17. UM-CAM	NO <sub>x</sub> , CO		Y	2.5

The response to perturbations in emissions of the indicated species were simulated by the models listing those species. Prescribed lifetime indicates that an additional simulation with an idealized prescribed lifetime tracer was also performed. Note that a few models did not perform all the regional perturbation experiments.

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**Table 2.** Mean (max, min) of total emissions in each region in Tg/yr across all models in their base run.

	S in SO <sub>2</sub>	BC	CO	N in NO <sub>x</sub>
East Asia	17 (21, 16)	1.9 (3.0, 1.5)	156 (220, 128)	7.0 (10.8, 5.2)
Europe	18 (25, 15)	1.0 (2.1, 0.7)	90 (130, 70)	8.4 (9.7, 7.2)
North America	11 (15, 10)	0.7 (1.1, 0.5)	129 (154, 107)	8.7 (9.4, 7.7)
South Asia	4 (5, 4)	0.9 (1.4, 0.7)	98 (145, 74)	3.3 (4.2, 2.6)

NO<sub>x</sub>=N+NO<sub>2</sub>

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**Table 3.** Annual average Arctic absolute mixing ratio changes due to 20% perturbations in anthropogenic emissions in each region.

	EA	EU	NA	SA
<i>Surface</i>				
Sulfate (pptm)	2.35±2.01 (13%)	12.4±9.8 (72%)	2.41±1.99 (14%)	0.25±0.25 (1%)
BC (pptm)	0.12±0.13 (14%)	0.66±0.68 (76%)	0.08±0.08 (9%)	0.01±0.01 (1%)
CO (ppbv)	2.23±1.07 (26%)	3.35±1.12 (39%)	2.42±0.75 (29%)	0.51±0.15 (6%)
Ozone (ppbv)	0.12±0.04 (27%)	0.11±0.07 (24%)	0.19±0.07 (42%)	0.03±0.01 (7%)
<i>500 hPa</i>				
Sulfate (pptm)	11.9±10.5 (26%)	22.8±20.4 (49%)	10.3±9.0 (22%)	1.51±1.82 (3%)
BC (pptm)	0.72±0.76 (36%)	0.84±0.94 (43%)	0.34±0.38 (17%)	0.08±0.10 (4%)
CO (ppbv)	2.38±1.01 (31%)	2.20±0.54 (28%)	2.52±0.71 (33%)	0.61±0.17 (8%)
Ozone (ppbv)	0.26±0.09 (23%)	0.35±0.11 (31%)	0.44±0.15 (40%)	0.07±0.05 (6%)
<i>250 hPa</i>				
Sulfate (pptm)	17.5±16.4 (36%)	14.3±14.2 (30%)	11.5±11.5 (24%)	4.92±5.27 (10%)
BC (pptm)	0.95±0.86 (46%)	0.44±0.41 (21%)	0.31±0.30 (15%)	0.37±0.41 (18%)
CO (ppbv)	1.35±0.48 (36%)	0.77±0.21 (20%)	1.11±0.28 (29%)	0.58±0.23 (16%)
Ozone (ppbv)	0.22±0.01 (25%)	0.22±0.17 (25%)	0.35±0.19 (39%)	0.10±0.07 (11%)

Values are positive, representing decreases with emission reduction. Ozone and sulfate changes are in response to NO<sub>x</sub> and SO<sub>2</sub> emissions changes, respectively. Values in parentheses are percentage of the total from these four source regions for each individual region. Values are multi-model means and standard deviations.

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**Table 4.** Annual average BC deposition to Greenland ( $\text{Tg}/\text{m}^2 \times 10^{-14}$ ) due to 20% of anthropogenic emissions from each region.

	EA	EU	NA	SA
<i>Greenland</i>				
All models	0.8±1.0	1.5±1.7	1.7±1.9	0.11±0.14
Excluding largest	0.5±0.4	1.2±1.4	1.2±1.2	0.06±0.04
Excluding smallest	0.9±1.0	1.7±1.7	1.9±1.9	0.12±0.15
<i>Arctic (excluding Greenland)</i>				
All models	24±30	88±96	15±17	2.4±4.1
Excluding largest	14±10	65±72	9.6±8.7	1.1±0.6
Excluding smallest	27±31	98±97	16±18	2.7±4.3

Values are multi-model means and standard deviations.

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**Table 5.** Lifetime or residence time (days) and correlation coefficients ( $R^2$ ) between those times and Arctic sensitivity across the models.

		EA	EU	NA	SA
CO subset (Global mean lifetime 62±12)	Surface correlation	.0	.3	.1	.8
	500 hPa correlation	.0	.0	.0	.8
	250 hPa correlation	.3	.3	.4	.6
CO all (Global mean lifetime 57±15)	Surface correlation	.3	.0	.3	.9
	500 hPa correlation	.4	.2	.3	.9
	250 hPa correlation	.4	.4	.5	.4
BC all	Mean residence time	4.5±1.6	5.6±1.2	4.8±1.3	6.6±1.6
	Surface correlation	.8	.8	.7	.4
	500 hPa correlation	.8	.6	.7	.4
	250 hPa correlation	.9	.5	.8	.5
SO <sub>4</sub> subset	Mean residence time	4.8±0.9	7.0±1.9	4.7±0.9	5.6±1.2
	Surface correlation	.2	.0	.5	.8
	500 hPa correlation	.2	.0	.1	.5
	250 hPa correlation	.2	.0	.3	.1
SO <sub>4</sub> all	Mean residence time	4.3±1.9	6.1±2.2	4.7±2.1	5.3±2.5
	Surface correlation	.2	.1	.1	.1
	500 hPa correlation	.5	.4	.5	.5
	250 hPa correlation	.6	.3	.7	.4

Global mean multimodel means and standard deviations of residence times for regional emissions for aerosols and global mean chemical lifetime from the control run for CO are given.  $R^2$  values are linear correlations between those times and the Arctic sensitivities at the given pressure levels. “All” and “subset” refer to the models used in the analysis (see text for subsets). The EMEP model was excluded since it includes the NH only and hence its global lifetime is not precisely equivalent to the others. In the CO analysis, GMI and MOZECH were not included due to problematic diagnostics.

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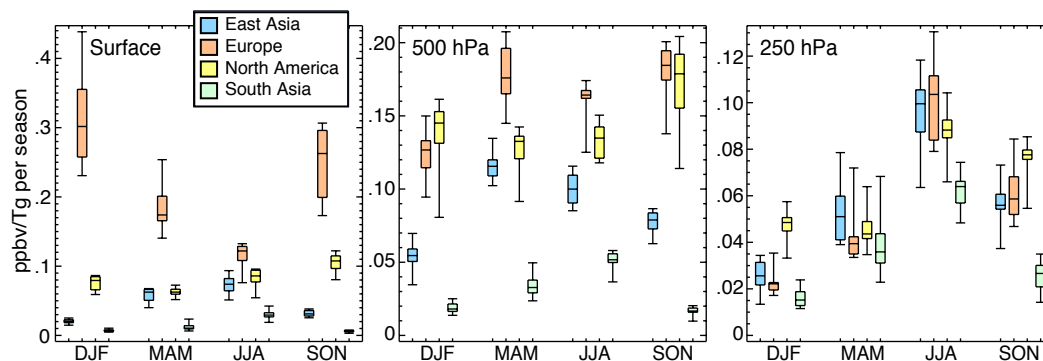


**Fig. 1.** The Arctic and the four source regions (shaded) used in this study.

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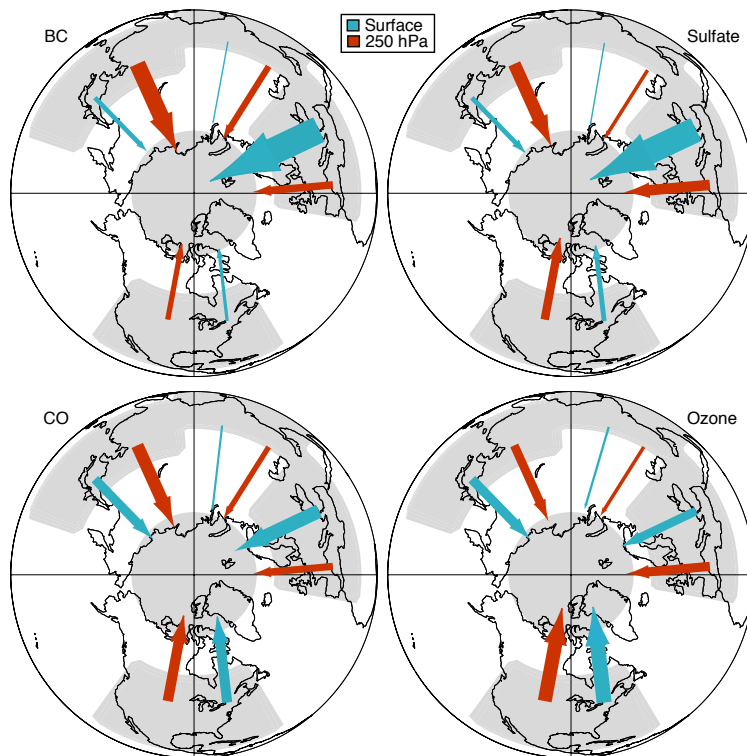
**Fig. 2.** Arctic sensitivity at three levels for the seasonal average CO-like tracer in terms of mixing ratio per unit emission from the given source region in the prescribed lifetime tracer simulations (10 models). Boxes show the central 50% of results with the median indicated by the horizontal line within the box, while the bars indicate the full range of model sensitivities.

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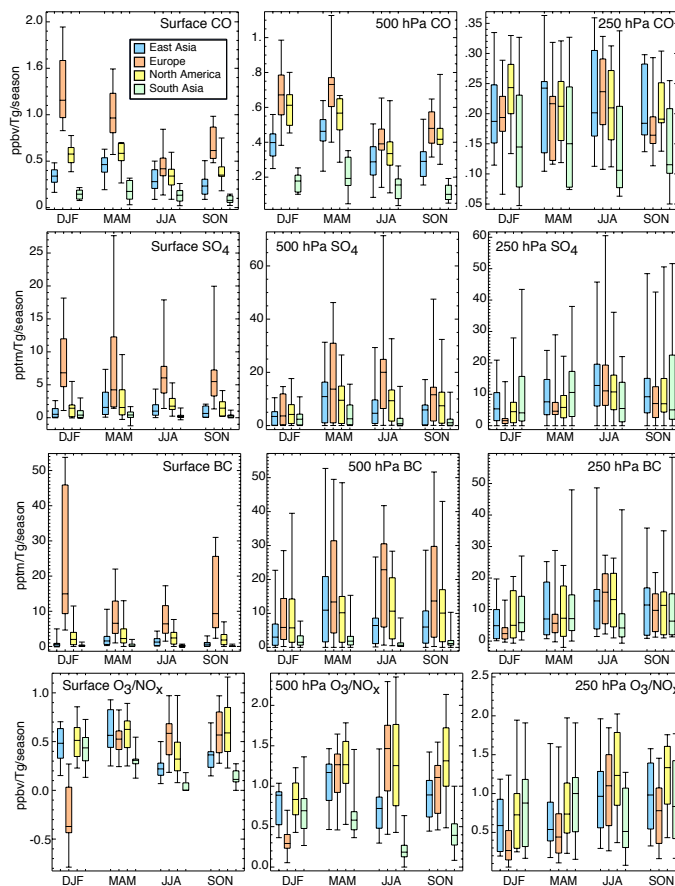


**Fig. 3.** Relative importance of different regions to annual mean Arctic concentration at the surface and in the upper troposphere (250 hPa) for the indicated species. Values are calculated from simulations of the response to 20% reduction in anthropogenic emissions of precursors from each region (using  $\text{NO}_x$  for ozone). Arrow width is proportional to the multimodel mean percentage contribution from each region to the total from these four source regions (as in Table 3).

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**Fig. 4.** Arctic sensitivity to emissions from the given region for seasonal averages of CO, sulfate, BC and ozone mixing ratios at the indicated heights. Sensitivities are the difference between the simulation perturbing a given emission and the control, normalized by the emissions change in the species (CO, BC) or its primary precursor (S in  $\text{SO}_2$  for  $\text{SO}_4$ , N in  $\text{NO}_x$  for  $\text{O}_3$ ) in the indicated source region. Symbols as in Fig. 2. Note change in vertical scale between columns.

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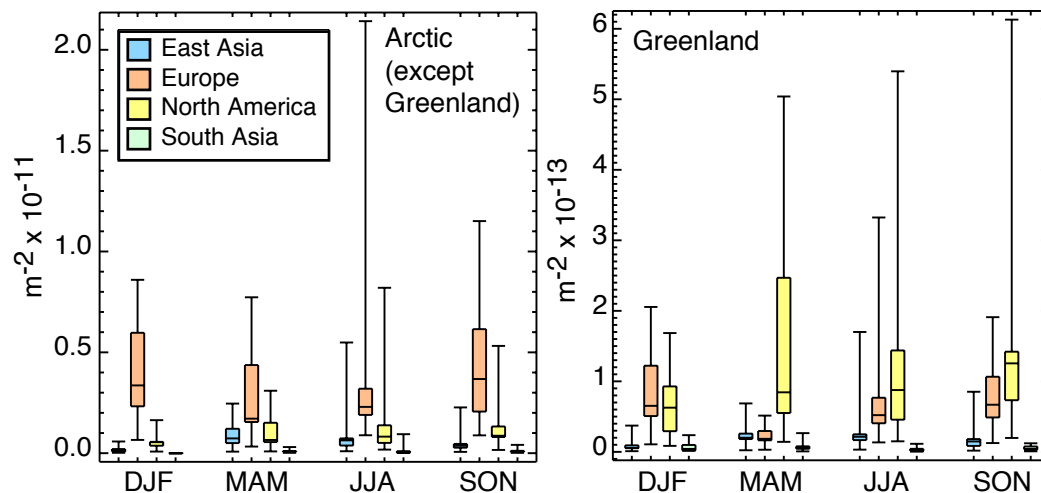
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**Fig. 5.** Sensitivity of Arctic-wide (left, excluding Greenland) and Greenland (right) BC deposition to regional emissions (Tg deposition per Tg emission per unit area per season). Values are calculated from the 20% anthropogenic emissions perturbation simulations.

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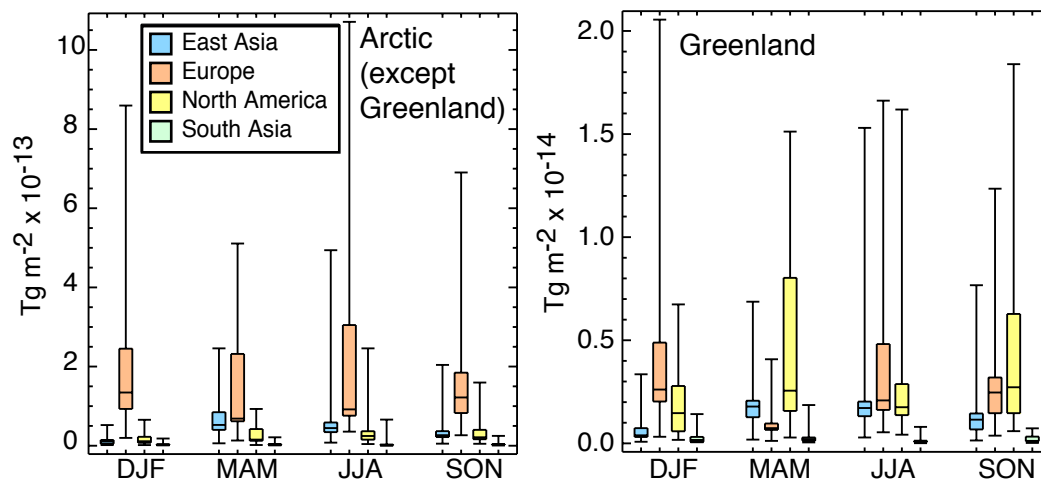
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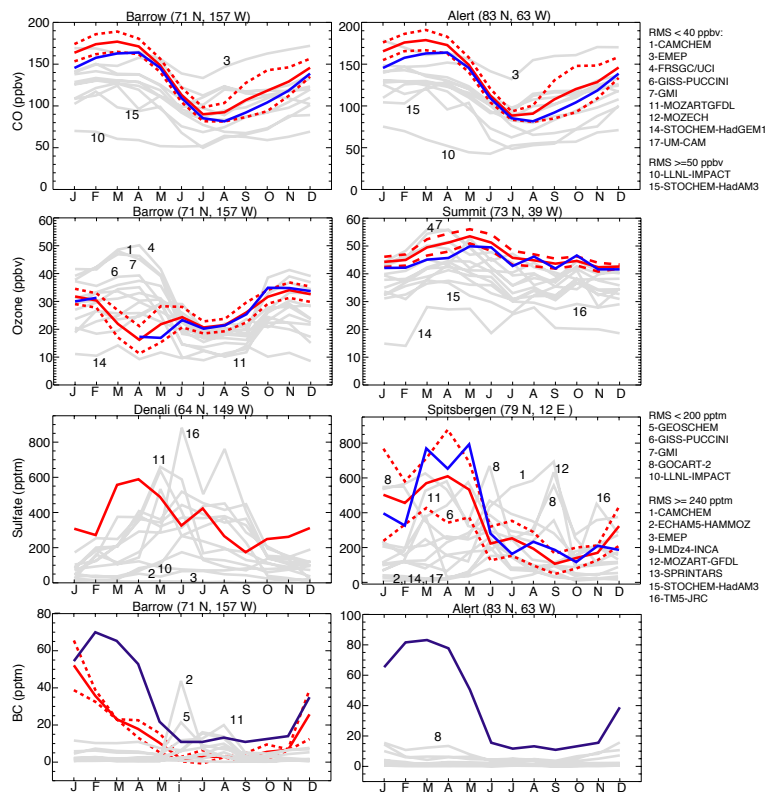


**Fig. 6.** Total Arctic-wide (left, excluding Greenland) and Greenland (right) BC deposition ( $T_g$  deposition per unit area per season) in response to 20% anthropogenic emission change from each source region.

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**Fig. 7.** Observed and modeled seasonal cycles of trace species surface concentrations at the indicated Arctic sites. Model results in all panels are in grey. Plots for CO (top row) and ozone (second row) show observations from the NOAA Global Monitoring Division, with 1992–2006 means and standard deviations in red (except for Summit O<sub>3</sub>, which is 2000–2006) and 2001 in blue. Sulfate plots (third row) show observations from the IMPROVE site at Denali during 1996–1999 (left) and from the EMEP site in Spitsbergen during 1999–2005 in red, with 2001 Spitsbergen data in blue. BC data (bottom row) are from the IMPROVE site at Barrow during 1996–1998 (red), and from Sharma et al. (2006) for both Barrow and Alert using equivalent BC over 1989–2003 (purple). Models are listed by RMS error scores to the right of each row using the groupings discussed in the text. Models that are separated from others are labeled with the numbers as in the text at right (or Table 1).

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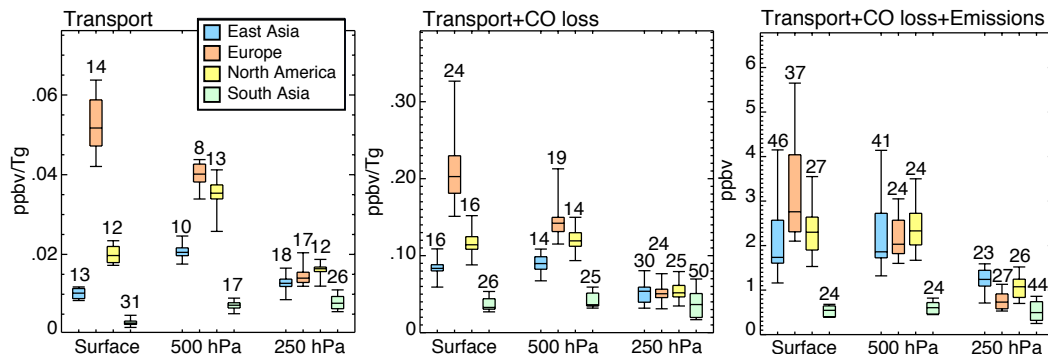
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**Fig. 8.** Annual Arctic average carbon monoxide response to source region emissions as a function of processes included in the models. The influence of transport is shown via the Arctic sensitivity in the prescribed lifetime CO-like tracer runs (left). The influence of transport plus CO oxidation is given by the sensitivity in the full chemistry run (center). The influence of transport plus oxidation plus emissions is also given by the full chemistry run, this time without normalization by the source region emissions change (right). Numerical values over each bar give the fractional variation (standard deviation as a percentage of the mean). Symbols as in Fig. 2.

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