

Using beryllium-7 to assess cross-tropopause transport in global models

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Abstract. We use the Global Modeling Initiative (GMI) modeling framework to assess the utility of cosmogenic beryllium-7 (^7Be), a natural aerosol tracer, for evaluating cross-tropopause transport in global models. The GMI chemical transport model (CTM) was used to simulate atmospheric ^7Be distributions using four different meteorological data sets (GEOS1-STRAT DAS, GISS II' GCM, fvGCM, and GEOS4-DAS), featuring significantly different stratosphere-troposphere exchange (STE) characteristics. The simulations were compared with the upper troposphere / lower stratosphere (UT/LS) ^7Be climatology constructed from ~25 years of aircraft and balloon data, as well as climatological records of surface concentrations and deposition fluxes. Comparison of the fraction of surface air of stratospheric origin estimated from the ^7Be simulations with observationally-derived estimates indicates excessive cross-tropopause transport at mid-latitudes in simulations using GEOS1-STRAT and at high latitudes using GISS II' meteorological data. These simulations also overestimate ^7Be deposition fluxes at mid-latitudes (GEOS1-STRAT) and at high latitudes (GISS II'), respectively. We show that excessive cross-tropopause transport of ^7Be corresponds to overestimated stratospheric contribution to tropospheric ozone. Our perspectives on STE in these meteorological fields based on ^7Be simulations are consistent with previous modeling studies of tropospheric ozone using the same meteorological fields. We conclude that the observational constraints for ^7Be and observed ^7Be total deposition fluxes can be used routinely as a first-order assessment of cross-tropopause transport in global models.

1 Introduction

Stratosphere-troposphere exchange (STE) of air masses and chemical species occurs at small-, synoptic and global-scales. It is typically associated with the occurrences of tropopause folding and cutoff cyclones and, more important, the global circulation of the atmosphere (Holton et al., 1995). While stratosphere-to-troposphere transport removes many chemical species from the stratosphere, it represents a significant source of ozone and other reactive species for the tropospheric chemical system (Stohl et al., 2003). Ozone is an important greenhouse gas, especially in the upper troposphere. It is a harmful pollutant near the surface where stratospheric ozone intrusions may make significant contributions (e.g., Lin et al., 2012, 2015; Langford et al., 2014). It is also the main precursor of hydroxyl radicals (OH) and thus plays an essential role in the oxidizing capacity of the troposphere. The stratosphere may make significant contributions to In a warmer climate, the stratosphere may increase its contribution to

1 tropospheric ozone levels due to a stronger residual circulation (Collin et al., 2003). Quantitative
2 understanding and prediction of anthropogenic (versus natural) perturbations to tropospheric
3 ozone require the use of global 3-D models; correctly representing the STE flux in these models
4 is therefore critical. However, current models show large (30%) uncertainty in predicted STE
5 fluxes of ozone (Stevenson et al., 2006). Here we use the Global Modeling Initiative (GMI)
6 modeling framework (Douglass, et al., 1999; Rotman et al., 2001) to assess the utility of the
7 aerosol tracer beryllium-7 (^7Be) for evaluating cross-tropopause transport in global models.

8 Beryllium-7 has a half-life of 53.3 days and is produced by cosmic ray spallation reactions in
9 the stratosphere and upper troposphere. After production, it attaches immediately to ubiquitous
10 submicron aerosols in the ambient air. The fate of ^7Be then becomes that of those aerosols, which
11 move with the air until scavenged by precipitation or deposited to the surface. ^7Be is a useful
12 aerosol tracer for testing wet deposition processes in a global 3-D model and is often used in
13 conjunction with the terrigenous ^{210}Pb aerosol tracer, as wet deposition is its principal sink and its
14 sources are relatively well known (e.g., Brost et al., 1991; Koch et al., 1996; Liu et al., 2001). On
15 the other hand, because of its source at high altitudes and the large concentration vertical
16 gradient, simulation of ^7Be tests the model's capability to describe stratosphere-to-troposphere
17 transport and subsidence in the troposphere (e.g., Liu et al., 2001; Allen et al., 2003).

18 Beryllium-7 has long been recognized as a tracer of downward transport from the
19 stratosphere to the troposphere (e.g., Husain et al., 1977; Viezee and Singh, 1980; Sanak et al.,
20 1985; Dibb et al., 1992, 1994; Rehfeld and Heimann, 1995). Husain et al. (1977) reported that
21 pulses of high ^7Be concentrations were often associated with air masses of stratospheric origin,
22 as indicated by large potential vorticity. Viezee and Singh (1980) showed that the ^7Be
23 concentrations over North America show strong positive correlations with the occurrence of
24 tropopause folding events over several latitude belts. ^7Be has also been combined with other
25 radionuclides (e.g., ^{10}Be , ^{90}Sr) as an indicator of transport of stratospheric air to the troposphere
26 (Raisbeck et al., 1981; Rehfeld and Heimann, 1995; Koch and Rind, 1998; Dibb et al., 1994;
27 Jordan et al., 2003; Zanis et al., 2003; Heikkila et al., 2008ab). Dutkiewicz and Husain (1985,
28 hereafter referred to as DH85) analyzed ^7Be and ^{90}Sr concentrations measured simultaneously in
29 samples from NASA's Global Atmospheric Sampling Program (GASP) and showed that on an
30 annual basis the stratosphere contributed ~25% of the observed ^7Be concentration at the northern
31 mid-latitude surface (~40% during late spring but only 10% during fall).

1 Beryllium-7 is also a useful tracer for vertical mixing and subsidence in the troposphere.
2 Feely et al. (1989) examined the factors that contribute to seasonal variations in ^7Be
3 concentrations in surface air. They found that the influences of variations both in the STE rate
4 and in the tropospheric vertical mixing rate are evident in concentrations at most sites in mid-
5 latitudes. Convective transport carries surface air upward and brings down the ^7Be at higher
6 altitudes to the surface layer. This is also reflected by the $^7\text{Be}/^{210}\text{Pb}$ ratio that peaks at the surface
7 in summer when convective activity is at its maximum (Koch et al., 1996). On the other hand,
8 despite the UT/LS source of ^7Be and the continental surface source of ^{222}Rn (precursor of ^{210}Pb),
9 ^7Be concentrations have been reported to be positively correlated with ^{210}Pb concentrations,
10 reflecting mixing of subsiding middle- and upper-tropospheric air with continental lower-
11 tropospheric air (Li et al., 2002; Dibb, 2007).

12 A number of observational studies have demonstrated the feasibility of using ^7Be to infer the
13 contribution of ozone-rich stratospheric air to ozone concentrations at ground level (e.g., Husain
14 et al., 1977; Tsutsumi et al., 1998; Helmig et al., 2007) and in the free troposphere (e.g., Johnson
15 and Viezee, 1981; Prospero et al., 1995; Graustein and Turekian, 1996; Kritz et al., 1991; Dibb et
16 al., 2003). These studies are usually based on the correlations between concurrent measurements
17 of ozone and ^7Be (as well as other tracers such as water vapor and calculated potential vorticity),
18 with positive ^7Be -ozone correlations indicating the presence of the upper-tropospheric or
19 stratospheric air. For instance, Helmig et al. (2007) showed a year-round correlation of ozone
20 with ^7Be at Summit, Greenland and concluded that surface-layer photochemical ozone
21 production does not appear to have a noticeable influence on surface ozone levels. However, it is
22 important to note that under some circumstances the observed positive correlations of surface
23 ozone with ^7Be may simply reflect the common vertical trends of tropospheric ^7Be and ozone
24 and does not necessarily indicate the influence of stratospheric air (Li et al., 2002). Recent global
25 modeling studies showed the models' capability to reproduce the observed ^7Be -ozone
26 relationships, providing useful constraints on the stratospheric (versus photochemical)
27 contribution to tropospheric ozone in the model (Li et al., 2002; Allen et al., 2003; Liu et al.,
28 2004).

29 Though correct representation of STE is essential for simulating ^7Be , ozone and other trace
30 species in the troposphere (e.g., Riese et al., 2012), large variations exist among models.
31 Stevenson et al. (2006) reported the average STE flux of ozone from 26 models of 552 ± 168
32 Tg/year. Observation-based estimates of STE fluxes of ozone into the troposphere are typically
33 in the range of 400-600 Tg/year (Murphy and Fahey, 1994). Some global models are able to

1 produce STE fluxes of ozone in this range (e.g., Olsen et al., 2004; Hsu et al., 2005; Hsu and
2 Prather, 2009; Lin et al., 2012; Young et al., 2013; Skerlak et al., 2014). For those models with
3 too fast (or rarely, too slow) cross-tropopause transport of ozone, one way to overcome the
4 difficulty is to use the Synoz (synthetic ozone) method (McLinden et al., 2000). The Synoz
5 method involves constraining the global mean cross-tropopause ozone flux to match a prescribed
6 value consistent with observations (e.g., Bey et al., 2001). But this method yields an unrealistic
7 stratospheric ozone field and therefore does not allow for on-line calculations of total ozone
8 columns and photolysis rates/heating rates (McLinden et al., 2000). By contrast, the other simple
9 model for stratospheric ozone (linearized ozone or Linoz) developed by McLinden et al. enables
10 these on-line calculations by linearizing the ozone tendency about the local ozone mixing ratio,
11 temperature, and the overhead column ozone density. Linoz is computationally efficient and can
12 be readily incorporated in climate models for long-term integrations. Nevertheless, using Linoz
13 (or full stratospheric chemistry) in global CTMs or chemistry-climate models that focus on the
14 troposphere requires a realistic model representation of net cross-tropopause total mass fluxes. In
15 this context, ^7Be tracer simulations may provide a simple way of evaluating cross-tropopause
16 transport in these models.

17 The intermodel differences in the estimated intensity and frequency of STE have been
18 attributed to different meteorological fields used to drive the models as well as different transport
19 algorithms and chemistry processes (Cristofanelli et al., 2003). The GMI modeling framework
20 facilitates the reduction of uncertainties of this kind. It is a modular CTM with the ability to
21 incorporate different inputs and components (e.g., meteorological fields, emission inventories,
22 chemical and microphysical mechanisms, and numerical schemes) that represent the different
23 approaches of current models. One of the distinct features of the GMI CTM is the ability to be
24 driven by different meteorological data sets (e.g., Douglass et al., 1999; Considine et al., 2005;
25 Liu et al., 2007) while maintaining the same algorithms for transport, deposition, emission,
26 chemistry and other pertinent processes. This allows us to isolate the uncertainties in the model
27 simulations due to differences in the meteorological data sets alone. The number of factors that
28 may contribute to differences in the simulations is thus reduced, as we previously showed using
29 the GMI simulated ^{222}Rn and ^{210}Pb radionuclide tracers (Considine et al., 2005).

30 In this paper, we present simulations of atmospheric ^7Be distributions with the GMI CTM
31 driven by four different meteorological data sets, including output from GEOS1-STRAT, GISS
32 II' GCM, fvGCM, and GEOS4-DAS, each featuring significantly different STE characteristics.
33 The reader is referred to **Table 1** for a list of acronyms of models and their driving

meteorological data sets. We use here not only the meteorological fields that are well known to have reasonably good representations of STE (e.g., fvGCM) but also those with poor representations (e.g., GEOS1-STRAT). The variability in simulated STE allows us to examine and assess the utility of ^7Be for evaluating STE in these (and other) global meteorological fields. We will illustrate the consequences of incorrect STE in terms of the simulation of tropospheric ^7Be and show that ^7Be concentrations and deposition fluxes may be used routinely as a first-order assessment for cross-tropopause transport in global models. We will discuss how the constraints on STE from ^7Be are consistent with previous modeling studies of tropospheric ozone using the same meteorological fields.

The remainder of this paper is organized as follows. Section 2 gives a brief description of the GMI model, ^7Be source and cross-tropopause flux, and ^7Be and ozone observational datasets used for evaluating the model. Section 3 evaluates model results with UT/LS and surface ^7Be data. Section 4 assesses cross-tropopause transport of ^7Be in different meteorological fields. Section 5 compares the results with previous modeling studies. Section 6 discusses the implications for the impact of STE on tropospheric ozone, followed by summary and conclusions in Section 7.

2 Model and data

2.1 GMI CTM

The GMI (<http://gmi.gsfc.nasa.gov>) CTM is a global 3-D composition model that includes a full treatment of both stratospheric and tropospheric photochemical and physical processes. It uses a 114-species chemical mechanism that combines the stratospheric mechanism of Douglass et al. (2004) with the tropospheric mechanism of Bey et al. (2001). The chemical mechanism includes both stratospheric and tropospheric heterogeneous reactions. Tropospheric aerosol (sulfate, dust, sea salt, organic carbon, and black carbon) fields are taken from the Goddard Chemistry, Aerosol, Radiation and Transport model (GOCART). Details of the model are described in Duncan et al. (2007, 2008), Strahan et al. (2007), and Considine et al. (2008). There is also a tropospheric version of the model that includes only tropospheric chemistry processes and uses the Synoz (synthetic ozone) scheme (McLinden et al., 2000) to ensure a given value for the total flux of ozone into the troposphere. The latter adopts a cross-tropopause ozone flux of about 530-590 Tg/year (Stevenson et al., 2006). In this study, we simulate ^7Be using the GMI CTM without chemistry, similar to the Considine et al. (2005) study that simulated the

radionuclides ^{222}Rn and ^{210}Pb . We use both the full-chemistry CTM and the tropospheric version of the model for ozone simulations.

The simulations presented in this paper differ only in the meteorological data used to drive the model. The four input meteorological data sets are from: (1). the Goddard Space Flight Center Data Assimilation Office (now Global Modeling and Assimilation Office or GMAO) GEOS1-STRAT data assimilation system (GEOS1-STRAT DAS, March 1997 - February 1998), (2). GISS II' GCM (Rind and Lerner, 1996), (3). the GMAO finite-volume GCM (fvGCM), and (4). GEOS4-DAS (February 2004 – January 2005). The GISS II' GCM data set is used for ^7Be simulations only. The two GCM data sets are intended to represent not any particular year but the contemporary climatological state of the Earth's atmosphere. Note that these data sets do not reflect the state-of-the-art, especially the first two. However, the choices are on purpose in order to see how a meteorological input with a poor representation of cross-tropopause transport affects the simulated tropospheric ^7Be . Vertical levels, top pressure, near-tropopause resolution, and bottom layer depth for each data set are listed in **Table 2**. The simulations presented here use a degraded horizontal resolution ($4^\circ \times 5^\circ$) for computational expediency. Degraded horizontal resolution slightly increases cross-tropopause transport (Liu et al., 2001). Nevertheless, our objective is to assess cross-tropopause transport in meteorological data sets at the resolution used to drive the model, not necessarily at the original or finer resolution.

The model uses the flux-form semi-Lagrangian advection scheme of Lin and Rood (1996) and a convective transport algorithm adapted from the CONVTRAN routine in the NCAR CCM3 physics package. The wet deposition scheme is that of Liu et al. (2001) and includes scavenging in wet convective updrafts, and first-order rainout and washout from both convective anvils and large-scale precipitation. The gravitational settling effect of cloud ice particles included in Liu et al. (2001) is not considered here. Dry deposition of ^7Be aerosols is computed using the resistance-in-series approach. The model tracks the bulk ^7Be aerosol mass. For ^7Be simulations, each simulation was run for six years, recycling the meteorological data for each year of the simulation; we use the sixth year output for analysis. For ozone simulations, the model was spun up for 10 years to remove the effect of initial conditions. Interannual variability in STE of ^7Be is not shown in this paper. However, model simulations driven by multi-year outputs from fvGCM (1994-1998) indicate that such interannual variability is much smaller than the differences due to using different meteorological data sets and does not affect the conclusions of this study.

The GMI CTM has been used previously to study the sensitivities of model simulations to different sets of meteorological input. Douglass et al. (1999) used chemical tracers in the GMI framework to assess three meteorological data sets, i.e., the NCAR Community Climate Model (CCM2), GEOS1-STRAT, and GISS II' GCM. They concluded that overall, CCM2 provides the best representation of the stratosphere. Considine et al. (2005) used the GMI model to simulate the radionuclides ^{222}Rn and ^{210}Pb using three different sets of meteorological inputs (GEOS1-STRAT, GISS II', and CCM3) to characterize the variability occurring in their simulations. Overall no simulation was found to be superior to the others when compared with the climatological observations of these radionuclides. The role played by convective transport and scavenging was found to differ substantially among the three meteorological data sets. Liu et al. (2007) analyzed and quantified the differences and uncertainties in GMI aerosol simulations solely due to different meteorological fields (GEOS1-STRAT, GISS II' GCM, and fvGCM). They suggested that the differences in the precipitation, convective mass flux, and horizontal advection from the three meteorological data sets explain much of the large discrepancies in the model-calculated aerosol concentrations.

2.2 Comparison of cloud and precipitation fields between meteorological data sets

Clouds and precipitation play a critical role in the transport and scavenging of ^7Be aerosols and thus in determining the lifetime, burden, and distribution of ^7Be in the troposphere. **Figure 1** and **Figure 2** compare the annual surface total precipitation and convective mass fluxes in the GEOS1-STRAT, GISS II' GCM, fvGCM and GEOS4-DAS meteorological data sets, respectively, following Liu et al. (2007). Also shown in **Figure 1** is the satellite climatology of surface total precipitation (1979-2009) from the Global Precipitation Climatology Project (GPCP) (Adler et al., 2003). The global mean precipitation rates are 1.9, 2.2, 2.6, 2.3 and 2.2 mm day⁻¹ for GEOS1-STRAT, GISS II' GCM, fvGCM, GEOS4-DAS and GPCP, respectively, with lightest precipitation in GEOS1-STRAT and heaviest in fvGCM. Compared to GPCP, GEOS1-STRAT and GEOS4-DAS significantly underestimate the precipitation in the mid-latitude storm track regions, while GISS II' GCM, fvGCM and GEOS4-DAS largely overestimate the observations in the tropics or subtropics. GISS II' GCM also underestimates the precipitation south of 50°S and north of 40°N. There are significant differences in the convective mass fluxes among the four meteorological data sets (**Figure 2**). Consistent with the precipitation, GEOS1-STRAT shows the weakest convection except in the tropical middle and upper troposphere, whereas fvGCM features the strongest convection in the boundary layer at 30-60°S. The effects

of the above differences in convection and precipitation between meteorological data sets on the results of this study will be examined through model sensitivity experiments.

2.3 ^7Be source

There is a large discrepancy in the published estimates of ^7Be production rates (Lal and Peters, 1967, referred to as LP67 hereafter; O'Brien et al., 1991; Masarik and Reedy, 1995; Masarik and Beer, 1999; Usoskin and Kovaltsov, 2008). Global mean column production rates over an average solar cycle range from $0.035 \text{ atoms cm}^{-2} \text{ s}^{-1}$ (Masarik and Beer, 1999), $0.063 \text{ atoms cm}^{-2} \text{ s}^{-1}$ (O'Brien et al., 1991), to $0.081 \text{ atoms cm}^{-2} \text{ s}^{-1}$ (LP67). The Masarik and Beer (1999) production function is smaller than other estimates by a factor of 2 or more. It may have underestimated the rate of ^7Be production and slightly overestimated changes in the production rate due to variations in geomagnetic and solar magnetic field strength (Koch et al., 2006; Field et al., 2006). We use in the model the LP67 source for 1958 (solar maximum year) since it leads to the best simulation of aircraft ^7Be observations in the stratosphere where ^7Be concentrations are mainly determined by a balance between production and radioactive decay (Koch et al., 1996; Liu et al., 2001). Koch et al. (1996) previously found that the O'Brien (1991) source yields model ^7Be concentrations near the surface and in the stratosphere that are much lower than observed. The rates of ^7Be production reported more recently by Usoskin and Kovaltsov (2008) broadly agree with those of LP67 with slightly (about 25%) lower global production rate and will be tested in a separate model study. The LP67 source is represented as a function of latitude and altitude (pressure) and does not vary with season (see Figure 1 of Koch et al., 1996). About 2/3 of atmospheric ^7Be is generated in the stratosphere and 1/3 in the troposphere. The ^7Be production rate correlates inversely with solar activity. At higher solar activity, cosmic rays are deflected away from the solar system and the ^7Be production rate is thus lower.

2.4 Constraint on stratospheric contribution to ^7Be at the surface

Cross-tropopause transport is important for simulating ^7Be in the troposphere. A useful constraint on the stratospheric contribution to tropospheric ^7Be is DH85's analysis of the observed $^7\text{Be}/^{90}\text{Sr}$ ratio in the stratosphere and ^{90}Sr concentrations at the surface. The presence of fissiogenic ^{90}Sr in the troposphere is due entirely to downward transport from the stratosphere, except for a few weeks right after a nuclear detonation. Both ^7Be and ^{90}Sr are associated with submicron particles; their fates during transport from the stratosphere are expected to be similar (no differential removal is expected). The stratospheric ^7Be component in surface air can therefore be determined as the product of the stratospheric $^7\text{Be}/^{90}\text{Sr}$ ratio and the surface ^{90}Sr concentration (DH85). By this procedure, DH85 showed that annually 23-27% (or about 25% on

average) of the ^7Be in surface air at northern mid-latitudes is of stratospheric origin. To use this constraint, we diagnose stratospheric contribution to ^7Be concentrations in the troposphere by transporting separately in the model the ^7Be produced in the stratosphere, as we previously applied in GEOS-Chem with GEOS1-DAS meteorological data (Liu et al., 2001). Since wet deposition removes both the stratospheric and tropospheric components of ^7Be at the same rate within each model gridbox, the diagnosed stratospheric fraction of ^7Be concentrations in the troposphere does not significantly depend on the rate of wet removal.

In the stratosphere, the production of ^7Be (source) is balanced by radioactive decay and net STE fluxes of ^7Be into the troposphere (sinks), i.e.,

$$\text{source } (^7\text{Be}) = \text{decay } (^7\text{Be}) + \text{STE } (^7\text{Be}). \quad (1)$$

Since the time scale for downward transport from the stratosphere to troposphere ($\sim 1\text{-}2$ years) is much longer than that for radioactive decay (half-life 53.3 days), the radioactive decay term is much larger than the STE flux term. Nevertheless, the STE term would become more important for a model atmosphere where STE is too fast. For the simulation of tropospheric (not stratospheric) ^7Be , the stratospheric influx to the troposphere may be adjusted by artificially scaling down (in the case of excessive STE) or up (in the case of too slow STE) the stratospheric ^7Be source. The extent to which ^7Be cross-tropopause transport is excessive or too slow in the model can be indicated by a scaling factor A, which is defined as the ratio of model to observed STE fluxes of ^7Be . We derive the scaling factor A as follows.

According to the DH85 constraint, we have for the observations

$$[^7\text{Be}]_{\text{T,G}} / [^7\text{Be}]_{\text{S,G}} = (1-0.25)/0.25 = 3 \quad (2)$$

where the left-hand side denotes the ratio of the tropospheric ($[^7\text{Be}]_{\text{T,G}}$) to stratospheric ($[^7\text{Be}]_{\text{S,G}}$) component of annual mean ^7Be concentrations in ground air at NH mid-latitudes. On the other hand, we have for a global model

$$[^7\text{Be}]_{\text{T,G}}' / [^7\text{Be}]_{\text{S,G}}' = (1-F)/F \quad (3)$$

where $[^7\text{Be}]_{\text{T,G}}'$ and $[^7\text{Be}]_{\text{S,G}}'$ are the *model* tropospheric and stratospheric components of annual mean ^7Be concentrations in surface air at NH mid-latitudes, respectively, and F is the corresponding fraction of surface air of stratospheric origin in the model. If the model reasonably represents the vertical transport and wet scavenging processes in the troposphere, we have

$$[^7\text{Be}]_{\text{T,G}}' = [^7\text{Be}]_{\text{T,G}} \quad (4)$$

Combining equations (2)-(4), we obtain the scaling factor

$$A \approx [^7\text{Be}]_{\text{S,G}}' / [^7\text{Be}]_{\text{S,G}} \approx 3F/(1-F). \quad (5)$$

We will discuss the sensitivity of F and A to the assumptions with respect to convective transport and scavenging processes in Section 4. The validity of equation (5) will also be evaluated with actual model calculations in that section. Unless otherwise specified, ^7Be cross-tropopause fluxes in the model calculations presented in this paper are not adjusted. However, we will use the scaling factor A as one of the metrics for comparing the STE characteristics of different meteorological data sets.

2.5 ^7Be and ozone observational data

^7Be . We estimate an average solar year value simply by averaging the long-term records of ^7Be observations multiplied by 0.72 to correct to the 1958 solar maximum source (Koch et al., 1996). The ^7Be deposition flux observations are from the compilation of Koch et al. (1996) and there are about 25 northern mid-latitude sites with available long-term ^7Be observations. The ^7Be surface concentration observations are from the data archive of the US Department of Energy (DOE) Environmental Measurements Laboratory (EML, now part of the Department of Homeland Security) Surface Air Sampling Program (SASP) beginning in the 1980's. We also use the long-term climatological data of ^7Be concentrations in the UT/LS constructed from ~25 years of aircraft and balloon observations. Between the late 1950s and the early 1980s, EML collected tropospheric and stratospheric aircraft and balloon measurements of numerous radionuclides as part of the DOE High Altitude Sampling Program (HASP). The data was compiled into a database in 1997 by R. Leifer and N. Chan of EML, called RADionuclide DATaBase (RANDAB). The reader is referred to Considine et al. (2005) for a brief description of the RANDAB database. This database is available at the Oak Ridge National laboratory's Carbon Dioxide Information Analysis Center (<http://cdiac.esd.ornl.gov/ndps/db1019.html>).

Ozone. We use tropospheric ozone column (TOC) determined with the tropospheric ozone residual method by subtracting measurements of MLS stratospheric column ozone (SCO) from OMI total column ozone (Ziemke et al., 2006; URL: http://acdb-ext.gsfc.nasa.gov/Data_services/cloud_slice) or using the TOMS and Solar Backscatter Ultraviolet (SBUV) combination (Fishman et al., 2003; URL: <http://science.larc.nasa.gov/TOR>). The OMI/MLS TOCs are from October 2004 - July 2008, and the TOMS/SBUV TOCs are from 1979-2005. We use climatological monthly average ozone profiles from 23 ozonesonde stations as constructed by Considine et al. (2008), based on Logan (1999) and Thompson et al. (2003). The ozonesonde data record is from 1985-2000 for extratropical stations, and from all available data prior to 2005 for tropical stations. The number of sondes at each station is adequate for

defining monthly means used to evaluate the accuracy of the model results (Considine et al., 2008). Surface ozone data are taken from Logan (1999).

3 Model evaluation with UT/LS and surface ^7Be data

In this section, we present model results of ^7Be simulations driven by four meteorological archives and evaluate them against long-term measurements at the surface and in the UT/LS. **Figure 3** shows the annual zonal mean concentrations (in units of millibecquerel per standard cubic meter or mBq/SCM^{-1}) of ^7Be in the four radionuclide simulations using GMI CTM. All four simulations overall show a similar pattern of tropospheric distribution. The highest concentrations are seen in the dry subsiding subtropics. Lowest ^7Be concentrations in surface air are found in the Southern Hemisphere mid-latitudes owing to scavenging by frequent large-scale precipitation (**Figure 1**). Low ^7Be concentrations are also associated with ITCZ, which is characterized by strong convergence and convective precipitation. It appears, however, that among all four simulations the GEOS1-STRAT simulation gives the highest concentrations in the subtropics and the GISS simulation shows the highest concentrations in the high latitudes. This is partly attributed to the differences in the latitudinal distribution of total precipitations in these meteorological archives (**Figure 1**).

Figure 4 compares four ^7Be simulations in the upper troposphere / lower stratosphere (UT/LS) with climatological distributions constructed from the ^7Be data contained in the RANDAB database, following Considine et al. (2005) who previously made a similar comparison for ^{210}Pb . Model output are sampled at the months, longitudes, latitudes, and altitudes of the ^7Be observations. **Figure 4a** compares the meridional distribution of ^7Be measurements made in the 12-16km altitude range with the four GMI simulations. **Figure 4b** shows the same comparison, but for the 16-20km altitude range. The 12-16km (about 200-100hPa) range lies within the upper troposphere in the tropics and the lower stratosphere at mid to high latitudes. The 16-20km (about 100-50hPa) range lies within the stratosphere at all latitudes.

At 12-16km (**Figure 4a**), the observations indicate comparatively low tropical upper tropospheric values of $\sim 35 \text{ mBq SCM}^{-1}$, with increasing trends toward high latitudes. The distribution is nearly symmetric about the equator, with more observations available in NH high latitudes. This latitudinal distribution of ^7Be concentrations reflects a larger production of ^7Be in the lower stratosphere at high latitudes and precipitation scavenging associated with deep convection in the tropics. All four simulations capture the observations at 12-16 km reasonably

1 well. The differences between the four simulated ^7Be concentrations are comparable or smaller
2 than the error limits.

3 At 16-20km (**Figure 4b**), the observations show a tropical minimum of $\sim 150 \text{ mBq SCM}^{-1}$,
4 with increasing concentrations toward high latitudes in both hemispheres. In the tropics and the
5 SH, the four ^7Be simulations indicate small differences. In the NH, the four ^7Be simulations
6 reveal large differences and bracket the observations. In particular, the GMI/GEOS1-STRAT
7 simulation gives the lowest ^7Be concentrations among the four simulations and is lower than the
8 observations. This appears to be due to excessive cross-tropopause transport in GEOS1-STRAT,
9 as further discussed below. On the other hand, as we will also discuss later, the fvGCM and
10 GEOS4-DAS meteorological fields have reasonable cross-tropopause transport. In the latter case,
11 stratospheric ^7Be concentrations are primarily determined by a balance between production and
12 radioactive decay in the stratosphere. Therefore the slightly overestimated ^7Be at 16-20km
13 suggests a slightly overestimated global production rate of ^7Be in the LP67 source. The Usoskin
14 and Kovaltsov (2008) source, which is about 25% lower than the LP67 source, would probably
15 yield better agreements with the ^7Be observations in the lower stratosphere.

16 **Figure 5a** compares the simulated and observed annual average concentrations of ^7Be near
17 the surface as a function of latitude. Observed data are from the EML SASP database and are
18 averaged into 10° latitude bins. Observations from sites with elevation higher than 500m are not
19 included because of uncertainties involved in sampling coarse-resolution models at high
20 elevation sites. Model results are sampled at observation locations and month. **Figure 5b** shows
21 the annual zonal mean surface ^7Be concentrations in the model to indicate the global
22 representativeness of the averages over the sampling sites. The observations indicate
23 concentration maxima in the subtropics associated with subsidence and minima in the tropics.
24 The tropical minimum reflects rapid scavenging within the ITCZ. Low ^7Be concentrations are
25 also observed at mid-latitudes due to efficient scavenging in the mid-latitude storm tracks.
26 Latitudinal trends (i.e., minima and maxima) of ^7Be concentrations are well simulated with all
27 meteorological fields except GISS II'. The GMI/GISS simulation shows too high ^7Be
28 concentrations at high latitudes; this is because of the well-known excessive cross-tropopause
29 transport at high latitudes in the GISS II' meteorological fields (e.g., Koch and Rind, 1998;
30 McLinden et al., 2000; Shindell et al., 2003). The overall positive biases in all simulations are
31 partly due to our correction of the long-term records of ^7Be observations (by a factor of 0.72) to
32 the 1958 solar maximum source (Section 2.5). We find that without this correction, the biases
33 would be significantly reduced.

Figure 5c compares the model-simulated annual mean total deposition fluxes of ^7Be at 25 northern mid-latitude sites from which long-term records of observations are available. The ^7Be deposition flux observations are from the compilation of Koch et al. (1996), previously used in Liu et al. (2001). The data from individual sites are averaged over 4° latitude bins. The model is sampled at observation locations. **Figure 5d** shows the annual zonal mean total deposition fluxes of ^7Be in the model to indicate the global representativeness of the sites. The observations show a maximum ($\sim 2100 \text{ Bq/m}^2/\text{yr}$) in the subtropics ($\sim 30^\circ\text{N}$) and the fluxes fall off with increasing latitude. The four ^7Be simulations show large discrepancies especially in the subtropics ($\sim 30^\circ\text{N}$). Overall, the GMI/fvGCM simulation agrees better with the magnitude of the observed fluxes while the GMI/GEOS4 simulation yields better latitudinal trends. GMI/GEOS4 simulates best the observations at the latitudes of 45°N - 60°N , but overestimates the observations by $\sim 50\%$ at 20°N - 40°N . The GMI/GISS simulation overestimates the observations at higher latitudes (45°N - 60°N) by a factor of ~ 2 . The GMI/GEOS1-STRAT simulation overestimates the observed ^7Be deposition fluxes at subtropical latitudes by up to a factor of 2.5 (30°N). As with the above model-observation comparison of surface ^7Be concentrations, the overall positive biases in model total deposition fluxes would be lower without the correction of ^7Be observations (by a factor of 0.72) to the 1958 solar maximum source (Section 2.5). However, we will show in the next section that these overestimated ^7Be deposition fluxes are largely due to model excessive cross-tropopause transport, especially with the GEOS1-STRAT and GISS II' meteorological fields.

4 Assessment of cross-tropopause transport of ^7Be in different meteorological archives

The above results indicate different levels of success with four meteorological fields in reproducing long-term records of surface and UT/LS ^7Be concentrations as well as total deposition fluxes. In this section, we quantify the contribution of ^7Be produced in the stratosphere to tropospheric ^7Be concentrations and deposition fluxes, followed by an assessment of cross-tropopause transport of ^7Be in the meteorological fields used.

Figure 6a shows the stratospheric fraction (%) of annual zonal mean atmospheric ^7Be concentrations (i.e., fraction of atmospheric ^7Be produced in the stratosphere) in the model simulations as a function of latitude and pressure. The fractions of significantly less than 100% in the lower stratosphere in all four simulations reflect mainly the seasonal movement of the tropopause. With GEOS1-STRAT, stratospheric contribution to lower-tropospheric ^7Be concentrations maximizes at 25 - 50°N (35-45%) and 25 - 40°S (30-35%). The tropical middle and

upper troposphere show the minimum in stratospheric impact (<30%). With GISS II', the stratospheric contribution to lower-tropospheric ^7Be concentrations peaks (30-40%) at southern high latitudes and remains nearly constant (30-35%) north of 30°N while it is quite small (<~10-20%) in the tropical middle and upper troposphere. The strong gradients in the subtropics suggest that the tropics are strongly isolated from the mid-latitudes in the GISS II' meteorological field. fvGCM and GEOS4-DAS show similar pattern of stratospheric influence on the troposphere; both indicate maximum contribution from stratosphere near 30-35°N (~25%) and 25-30°S (~20-25%) in the lower troposphere. However, GEOS4-DAS shows larger contributions from the stratosphere to the troposphere (especially the free troposphere) than fvGCM does by a few percent, consistent with the overestimated deposition fluxes at 20°N-40°N by GEOS4-DAS (**Figure 5c**). The area of minimal stratospheric influence in the tropics is also narrower in GEOS4-DAS.

Figure 6b shows the stratospheric fraction (%) of annual zonal mean surface ^7Be concentrations and that of annual zonal mean ^7Be total deposition fluxes ($\text{Bq}/\text{m}^2/\text{yr}$) in the model simulations as a function of latitude. With all meteorological fields except GISS II', maximum stratospheric contribution to total deposition fluxes (versus surface ^7Be concentrations) is shifted toward higher latitudes, reflecting scavenging by frequent mid-latitude precipitation and the dry subsidence in the subtropics. Stratospheric fractions of surface ^7Be concentrations at NH mid-latitude are about 38% (GEOS1-STRAT), 33% (GISS II'), and 23-24% (fvGCM and GEOS4-DAS). As discussed in Section 2.4, the observed $^7\text{Be}/^{90}\text{Sr}$ ratio suggests that 23-27% of the ^7Be in surface air at northern mid-latitudes is of stratospheric origin (DH85). According to this constraint, cross-tropopause transport of ^7Be and subsequent transport to the surface in the GEOS1-STRAT and GISS II' meteorological fields is excessive. On the other hand, it should be noted that the fvGCM and GEOS4-DAS simulations show results remarkably consistent with the DH85 constraint, suggesting that stratospheric influences on surface ^7Be concentrations in these two meteorological fields are reasonable. However, DH85 did not provide constraints on latitudinal variation of stratospheric influence on surface ^7Be . Of the four meteorological fields, GEOS1-STRAT, fvGCM and GEOS4-DAS show very similar latitudinal distribution of stratospheric influence at the surface (i.e., peak in the subtropics and valley in the tropics or polar regions). By contrast, GISS II' shows the largest impact of the stratosphere at high latitudes.

Similarly, as shown above, the model overestimates the long-term records of ^7Be deposition flux observations at mid-latitudes (and subtropics) with GEOS1-STRAT and at high latitudes with GISS II' (**Figure 5c**). Interestingly, the fvGCM (and to a lesser extent GEOS4-DAS)

simulation yields ^7Be deposition fluxes close to the observations. This suggests that the DH85 constraint and observed ^7Be deposition fluxes are two complementary constraints on cross-tropopause transport of ^7Be . We therefore use the DH85 constraint to assess the cross-tropopause transport of ^7Be in the meteorological fields.

Using the approach described in Section 2.4 (i.e., reduced cross-tropopause transport flux by artificially scaling down the stratospheric ^7Be source in the simulation of tropospheric ^7Be), we determine the scaling factors for GEOS1-STRAT and GISS to be 1.92 and 1.35, respectively. With the adjustment of ^7Be cross-tropopause fluxes for GEOS1-STRAT and GISS, the model calculated stratospheric fraction of ^7Be concentrations in surface air at NH mid-latitudes are indeed close to 25% (i.e., agree with the DH85 constraint) (**Figure 7**), thus supporting the validity of equation (5). With the adjustment, some simulations also simulate better surface ^7Be concentrations and total deposition fluxes at the subtropics (GEOS1-STRAT) and at high latitudes (GISS II') (**Figure 8** vs. **Figure 5**). The improvement is clearer for total deposition fluxes than for surface concentrations. As discussed below, on a global scale total deposition fluxes are sensitive to STE fluxes of ^7Be into the troposphere, while surface concentrations are principally dependent on the overall wet removal rate.

Table 3 shows the annual average global budgets of tropospheric ^7Be in the four GMI simulations. With an adjustment of ^7Be cross-tropopause fluxes, the global burdens and residence times of tropospheric ^7Be in GMI/GEOS1-STRAT and GMI/GISS are reduced. In GMI/GEOS1-STRAT the source and sink terms become much closer to that in fvGCM and GEOS4-DAS. A reduction of global ^7Be STE fluxes of 0.04 g d^{-1} results in a decrease of total deposition fluxes of 0.03 g d^{-1} and radioactive decay of 0.01 g d^{-1} . In GMI/GISS the changes in the budget terms are relatively small due to the smaller adjustment of ^7Be cross-tropopause fluxes. Nevertheless, a reduction of global ^7Be STE fluxes of 0.01 g d^{-1} results in a decrease of total deposition fluxes of 0.01 g d^{-1} . These calculations indicate that globally the ^7Be total deposition fluxes are sensitive to STE fluxes of ^7Be into the troposphere.

The model calculated stratospheric fraction of ^7Be in the troposphere may be sensitive to the model diagnosed location of the tropopause, for which there is some uncertainty. For instance, Stajner et al. (2008) used four different definitions of the tropopause on the basis of temperature lapse rate (World Meteorological Organization or WMO definition), potential vorticity (PV), and isentropic surfaces or ozone surfaces. They found that the WMO tropopause was about 0.7-1 km (in the northern mid-latitude) or 0.5-1 km (in the tropics) higher than the ozone or PV determined tropopause. We examine the sensitivity of model diagnosed stratospheric fraction of

1 tropospheric ^7Be concentrations to the location of tropopause (not shown) by lowering
2 tropopause height by one model level (approximately 1.2 km, 1.7 km, 1.1 km, and 1.1 km for
3 GEOS1-STRAT, GISS II', fvGCM and GEOS4-DAS, respectively). Results indicate that
4 stratospheric fractions of surface ^7Be concentrations increase by 5-10%, thus requiring larger
5 adjustments of cross-tropopause transport of ^7Be in the meteorological fields in order to meet the
6 DH85 constraint. This also suggests that using the DH85 constraint requires relatively high
7 vertical resolution near tropopause in the model.

8 While the model diagnosed stratospheric fraction of tropospheric ^7Be concentrations is
9 mainly determined by the STE processes in the UT/LS, it may also be sensitive to precipitation
10 scavenging and convective transport in the troposphere. **Figure 9** shows the latitude-pressure
11 cross sections of the differences in the stratospheric fraction (%) of annual zonal mean
12 tropospheric ^7Be concentrations between the standard simulation and a simulation where
13 precipitation scavenging is turned off. Also shown are the corresponding differences near the
14 surface. The stratospheric fraction of tropospheric ^7Be is found to be only weakly dependent on
15 precipitation scavenging, with <5% change in most of the troposphere and <2.5% change near
16 the mid-latitude surface. **Figure 10** shows a similar plot, except that convective transport and
17 scavenging are turned off in the sensitivity simulation. Similarly, the stratospheric fraction of
18 tropospheric ^7Be is not sensitive to convective transport and scavenging processes, with <1%
19 changes near the mid-latitude surface.

21 5 Comparison with previous modeling studies

22 In this section we compare the GMI CTM results for cross-tropopause transport of ^7Be with
23 previous modeling studies based on the same or similar meteorological fields.

24 Liu et al. (2001) found that STE flux of ^7Be was overestimated with the GEOS1-STRAT
25 fields in the GEOS-Chem model, consistent with this study using GMI CTM. However, Liu et al.
26 (2001) found that the reduction required to match the DH85 constraint is a factor of 3.5 for the
27 GEOS1-STRAT archive with $4^\circ \times 5^\circ$ resolution, compared to a factor of 2.5 in the present study.
28 The larger reduction in the former reflects the inclusion of ice particle gravitational settling
29 effect, which results in increased transport from the upper to lower troposphere, as well as the
30 inclusion of the diagnosed tropopause model layer as part of the stratosphere (versus the
31 troposphere). Interestingly, when specifying ozone concentrations in the lower stratosphere
32 (70hPa) and letting the model (GEOS-Chem) transport this ozone as an inert tracer into the
33 troposphere, Bey et al. (2001) found a similar overestimate in an ozone simulation with the

1 GEOS-1 data, as diagnosed by the simulation of tropospheric ozone concentrations at high
2 latitudes in winter where transport from the stratosphere is a major source. This indicates that
3 the simulation's deficiency in cross-tropopause transport as diagnosed using ^7Be tracers has
4 similar consequences for cross-tropopause transport of ozone.

5 Koch and Rind (1998) used a 31-layer version of the GISS GCM to simulate ^7Be and ^{10}Be
6 and used tropospheric $^{10}\text{Be}/^7\text{Be}$ as indicator of STE. Based on limited observations, they
7 suggested that leakage into the troposphere is somewhat excessive in the model, particularly at
8 high latitudes. Using the GISS II' GCM, McLinden et al. (2000) found that a large fraction of the
9 cross-tropopause transport of ozone occurs at the poles which is inconsistent with the current
10 understanding of stratosphere-troposphere exchange, despite that the global stratosphere-
11 troposphere exchange fluxes of ozone compare well with their best estimate of 475 ± 120 Tg/year
12 based on measurements and tracer-tracer correlation. Shindell et al. (2003) presented an updated
13 version of the GISS II' climate model which still overestimates ozone in the middle troposphere
14 at high latitudes, likely reflecting deficiencies in the model's downward transport of stratospheric
15 air. Our conclusions about cross-tropopause transport of ^7Be in GISS II' in this work are
16 consistent with these previous studies. Overestimated STE fluxes of ^7Be as diagnosed in
17 GMI/GISS based on the DH85 constraint simply reflect the incorrect latitudinal distribution of
18 cross-tropopause transport, that is, too fast STE at higher latitudes and too slow STE at lower
19 latitudes. The DH85 constraint was only applicable and applied for NH mid-latitude surface and
20 thus does not provide constraint on the model global STE flux of ^7Be if the latitudinal
21 distribution of STE is incorrect.

22 The large-scale stratospheric transport (Brewer-Dobson circulation) in fvGCM has been
23 shown to be realistic (Douglass et al., 2003) and mean age of stratospheric air is similar to
24 observations (Strahan and Douglass, 2004; Douglass et al., 2008; Strahan et al., 2009). This
25 suggests credible cross-tropopause transport of mass and ozone in fvGCM because the large-
26 scale exchange between the stratosphere and troposphere is largely tied to the Brewer-Dobson
27 circulation through the overworld wave driving (Holton et al., 1995; Olsen et al., 2004). Based
28 on this finding, the meteorological data from fvGCM was used to drive GMI CTM by several
29 authors to study tropospheric ozone. Considine et al. (2008) evaluated near-tropopause ozone
30 distributions with ozonesonde data. Terao et al. (2008) examined the role of variability in the
31 input of stratospheric ozone on the interannual variability of tropospheric ozone in the northern
32 extratropics. Liang et al. (2009) investigated the impact of stratosphere-to-troposphere transport
33 on tropospheric ozone and NO_x chemistry over the Arctic. By contrast, GEOS4-DAS tends to

1 have too strong of a residual circulation, and the age of air is too young as compared to
2 observations (Schoeberl et al., 2003; Schoeberl, 2004; Douglass et al., 2008). A GMI CTM
3 simulation driven with the GEOS4-DAS meteorological fields showed the model's inadequacy
4 in simulating upper-tropospheric ozone (Liang et al., 2009). These findings are consistent with
5 what we illustrated in this study from a perspective of ^7Be tracers. That is, GEOS4-DAS features
6 larger impact of STE on the troposphere (especially UT) than fvGCM does, while the latter has
7 more credible cross-tropopause transport as constrained by observed ^7Be deposition fluxes
8 (**Figure 5c**) and the DH85 criterion (**Figure 6**).

10 **6 Implications for cross-tropopause transport of ozone**

11 In this section we discuss the implications of different characteristics of cross-tropopause
12 transport of ^7Be for stratospheric influence on tropospheric ozone in different meteorological
13 fields. At the time of this study, the GMI full-chemistry model can be driven with GEOS1-
14 STRAT, fvGCM and GEOS4-DAS, but not GISS II' meteorological fields. This allows us to
15 examine any potential relationship between the cross-tropopause transport of ^7Be and ozone
16 when these fields are used to drive the model.

17 Ozonesonde, surface and satellite observations provide useful constraints on the stratospheric
18 contribution to tropospheric ozone (e.g., Rind et al., 2007; Lin et al., 2012). **Figure 11** shows
19 comparisons of model tropospheric ozone profiles with annual mean ozonesonde observations
20 for a range of latitudes (Considine et al., 2008). These results are typical of other stations at
21 similar latitudes. The GMI/GEOS1-STRAT simulation produces excessive ozone throughout the
22 troposphere at all latitudes except in the tropics while the GMI/fvGCM and GMI/GEOS4-DAS
23 simulations are generally in agreement with the observations (with slightly overpredicted ozone
24 in the mid-latitude upper troposphere). The GEOS1-STRAT simulation has the largest
25 overestimate of O_3 in spring. We also compared model surface ozone concentrations with the
26 Logan (1999) surface ozone dataset (not shown). Among the three GMI simulations, the
27 GMI/GEOS1-STRAT simulation shows the largest errors in surface ozone concentrations during
28 winter and spring when stratospheric contribution is at its peak. These are in line with the relative
29 magnitudes of cross-tropopause transport efficiencies of ^7Be in the three meteorological fields
30 (i.e., too fast STE in GEOS1-STRAT), discussed in previous sections. Indeed, the tropospheric
31 version of the GMI/GEOS1-STRAT model with constrained STE flux of ozone using the Synoz
32 approach (about 579 Tg/year) simulates ozonesonde observations of tropospheric ozone
33 reasonably well (dotted line, **Figure 11**).

Figure 12 shows GMI simulated annual zonal mean tropospheric ozone column (TOC), in Dobson Units, compared with observed climatologies from TOMS/SBUV (1979-2005; Fishman et al., 2003) and OMI/MLS (October 2004 - July 2008; Ziemke et al., 2006). The WMO definition of thermal tropopause is used to calculate the model TOC. While the GMI/fvGCM and GMI/GEOS4-DAS simulations are similar and overestimate TOC by up to ~20 DU, the GMI/GEOS1-STRAT simulation overestimates TOC by as much as ~40 DU. The excessive O₃ in the GMI/GEOS1-STRAT simulation with maxima at 30°N and 30°S suggests downward transport of ozone from the stratosphere is too fast. The tropospheric version of the GMI/GEOS1-STRAT model with constrained STE flux of ozone provides a much better simulation of global TOCs (red dashed line, **Figure 12**), which are comparable to those from GMI/fvGCM and GMI/GEOS4-DAS simulations. However, model TOCs are still ~10-14DU larger than satellite observations in the subtropics and mid-latitudes. Previously, Ziemke et al. (2006) considered uncertainties in both model and observations and subjectively interpreted model-OMI/MLS TOC differences of 10 DU and higher as significant. As Stajner et al. (2008) noted, a low extratropical tropopause used by Ziemke et al. (2006) may have played an important role in the underestimation of OMI/MLS TOC. Yang et al. (2010) also found that their OMI/MLS potential vorticity mapped TOCs are smaller than ozonesonde TOCs by 5.9 DU with a standard deviation of the differences of 8.4 DU. On the other hand, the GMI/fvGCM simulation tends to overestimate ozone just below the tropopause at mid-latitudes (**Figure 11**); these biases do not appear to be due to excessive stratospheric influence (Considine et al., 2008). Current global models also tend to overpredict surface ozone during summer and early fall over the eastern U.S. and Japan (Fiore et al., 2009). Therefore the simulated TOCs are very likely biased high.

We further examine the relationship between the cross-tropopause transport of ⁷Be and ozone with the GEOS1-STRAT meteorological fields, in which case STE is known to be too fast. **Figure 13a** shows the latitudinal variations of annual zonal mean tropospheric ⁷Be column overestimate ($\Delta^7\text{Be}$) and TOC overestimate (ΔTOC) in the GMI/GEOS1-STRAT simulation. $\Delta^7\text{Be}$ is obtained by subtraction of the STE-flux-adjusted simulation (Section 2.4) from the standard simulation. ΔTOC is obtained by subtracting the GMI tropospheric model simulation (with STE flux of ozone about 579 Tg/year) from the GMI full-chemistry model simulation. **Figure 13b** shows the correlation between the global distributions of $\Delta^7\text{Be}$ and ΔTOC . The lines of best fit are calculated using the reduced-major-axis (RMA) method (Hirsch and Gilroy, 1984). Standard errors for the intercept and the slope are computed as described by Miller and Kahn

(1962). Overall, the location of overestimated ozone follows that of overestimated ^7Be , with both maxima near 30°N and 30°S . The strong correlation between $\Delta^7\text{Be}$ and ΔTOC implies that ^7Be is a good indicator of cross-tropopause transport of ozone. These support our conclusion that ^7Be is a useful utility for assessing cross-tropopause transport of ozone in global models.

7 Summary and conclusions

We have assessed the ability of the Global Modeling Initiative (GMI) chemical transport model (CTM) using different meteorological data sets to simulate the atmospheric distributions of ^7Be , a natural aerosol tracer originating from the upper troposphere/lower stratosphere and removed from the troposphere primarily by wet deposition. The model was driven by four meteorological data sets (GEOS1-STRAT, GISS II', fvGCM, GEOS4-DAS) which feature significantly different cross-tropopause transport characteristics. The GMI modeling framework was configured such that the variability between the simulations mainly reflects the use of different meteorological data. Our goal was to assess the utility of ^7Be as a tracer of cross-tropopause transport in global models and develop a methodology to exploit such a utility. We have also discussed the implications of excessive cross-tropopause transport as revealed by ^7Be simulations for the modeling of tropospheric ozone.

We evaluated the four simulations of ^7Be with RANDAB, a unique database of upper atmosphere radionuclide climatological observations compiled by the DOE (now DHS) Environmental Measurement Laboratory, as well as long-term measurements at the surface. Model simulations capture the UT/LS observations with respect to latitudes. The GMI/GEOS1-STRAT simulation shows the lowest ^7Be concentrations among the four simulations in the lower stratosphere, and underestimates the observations. This reflects the well-known highly overestimated cross-tropopause transport in GEOS1-STRAT DAS. At the surface, GMI/GISS II' reproduces the observed latitudinal trends of ^7Be concentrations, but shows too high concentrations at high latitudes. The GMI/fvGCM simulated ^7Be deposition fluxes are the closest to the observations, while the GMI/GEOS1-STRAT overestimates the observed ^7Be deposition fluxes at subtropical latitudes by up to a factor of 2.5 (30°N) and the GMI/GISS simulations at high latitudes ($45\text{--}60^\circ\text{N}$) are too high by a factor of 2. We were able to show that the observed ^7Be deposition fluxes offer a strong constraint on stratosphere-to-troposphere transport in global models.

We examined the observational constraint from Dutkiewicz and Husain (1985) (DH85) on the stratospheric contribution to tropospheric ^7Be using the GMI modeling framework. DH85

analyzed the observed $^7\text{Be}/^{90}\text{Sr}$ ratio, which suggests that 23-27% of the ^7Be in surface air at northern mid-latitudes is of stratospheric origin. This constraint offers a sensitive test of cross-tropopause transport in global models. Comparison of the fraction of surface air of stratospheric origin estimated from the ^7Be simulations with the DH85 constraint indicates excessive cross-tropopause transport at mid-latitudes with the GEOS1-STRAT meteorological fields and at high latitudes with the GISS II' fields. Interestingly, these simulations also overestimate observed ^7Be deposition fluxes at middle and high latitudes, respectively. With a correction to cross-tropopause flux, the model simulates better surface ^7Be concentrations and total deposition fluxes. By contrast, the fvGCM meteorological data yield the most reasonable cross-tropopause transport of ^7Be according to the DH85 constraint, consistent with the fact that the GMI/fvGCM simulated ^7Be deposition fluxes are closest to the observations. These results illustrate that the GMI framework is very useful for characterizing and helping reduce uncertainties in the processes such as cross-tropopause transport in the meteorological fields that are used to drive chemical transport models. Note that since wet deposition removes both the stratospheric and tropospheric components of ^7Be nondiscriminatively, the model diagnosed fraction of ^7Be of stratospheric origin does not significantly depend on the rate of wet removal.

The model diagnosed stratospheric fraction of ^7Be in surface air is sensitive to the diagnosed location of tropopause, in particular when the model vertical resolution is relatively coarse (>1 - 1.5km) near the tropopause region. This suggests that stratospheric fraction of ^7Be is a more useful diagnostic when the model has sufficient vertical resolution (<1 - 1.5km) so that the tropopause can be well defined. We used the WMO definition of thermal tropopause and include the diagnosed tropopause model layer as part of the troposphere (versus the stratosphere). As such our assessment of cross-tropopause transport of ^7Be in the four meteorological data sets in the GMI CTM is consistent with previous modeling studies of stratospheric influence on tropospheric ozone.

Incorrect cross-tropopause transport of ^7Be implies misrepresented downward influx of stratospheric ozone to the troposphere in a model. We demonstrated this by examining the relationship between the cross-tropopause transport of ^7Be and ozone as simulated by GMI CTM driven with GEOS1-STRAT, fvGCM and GEOS4-DAS meteorological fields. We found that excessive cross-tropopause transport of ^7Be corresponds to overestimated stratospheric contribution to tropospheric ozone, as constrained by ozonesonde, surface and satellite observations.

1 In summary, the ^7Be simulation, which is computationally cheap and technically simple, in
2 combination with the DH85 ^7Be observational constraint and observed ^7Be deposition fluxes
3 may be used routinely to assess cross-tropopause transport in global models. We recommend
4 separate transport of the ^7Be produced in the stratosphere (^7Be -strat) to evaluate the ratio of ^7Be -
5 strat to total ^7Be (i.e., beryllium-7 produced in both the stratosphere and the troposphere) in
6 surface air against the DH85 constraint. This can serve as a first-order assessment of cross-
7 tropopause transport in the meteorological fields. With improved estimates of ^7Be production
8 rates as well as their year-to-year variations, model multi-year ^7Be simulations together with
9 long-term observations would provide useful constraints on the interannual variability of STE.
10 While this study uses ^7Be alone, future modeling work will include using $^{10}\text{Be}/^7\text{Be}$, a more
11 sensitive indicator of STE (Rehfeld and Heimann, 1995; Koch and Rind, 1998; Jordan et al.,
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3 Table 1. Acronyms of the model and driving meteorological data sets
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Model / Data Set	Acronym
GMI CTM	Global Modeling Initiative Chemical Transport Model
GEOS1-STRAT DAS	Goddard Earth Observing System Data Assimilation System – version 1 in support of the Stratospheric Tracers of Atmospheric Transport mission
GISS II’ GCM	Goddard Institute for Space Studies General Circulation Model – version II’
fvGCM	Global Modeling and Assimilation Office (GMAO) finite-volume GCM
GEOS4-DAS	GEOS Data Assimilation System– version 4

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1 Table 2. Characteristics of meteorological data sets used to drive the GMI CTM

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Data Set	Number of levels	Top Pressure (hPa)	Vertical Coordinate	Interface Pressure (hPa) ^a	Near-Tropopause Resolution (km)	Bottom layer depth (hPa, m)	Update Period (h)
GEOS1-STRAT	46	0.1	σ	N/A	~1.0	~12.13hPa, ~100m	6
GISS II'	23	0.002	σ -P	150	~1.8-2.5	~24.46hPa, ~200m	3
fvGCM	42 (55 ^b)	0.9 (0.01 ^b)	σ -P	200	~1.0	~14.89hPa, ~130m	3
GEOS4	42 (55 ^b)	0.9 (0.01 ^b)	σ -P	200	~1.0	~14.89hPa, ~130m	3

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4 ^aThe hybrid vertical coordinate consists of sigma (σ) levels below the interface pressure and constant pressure (P) levels above.

5 ^bThe total number of vertical levels and top level pressure in the original meteorological data set.

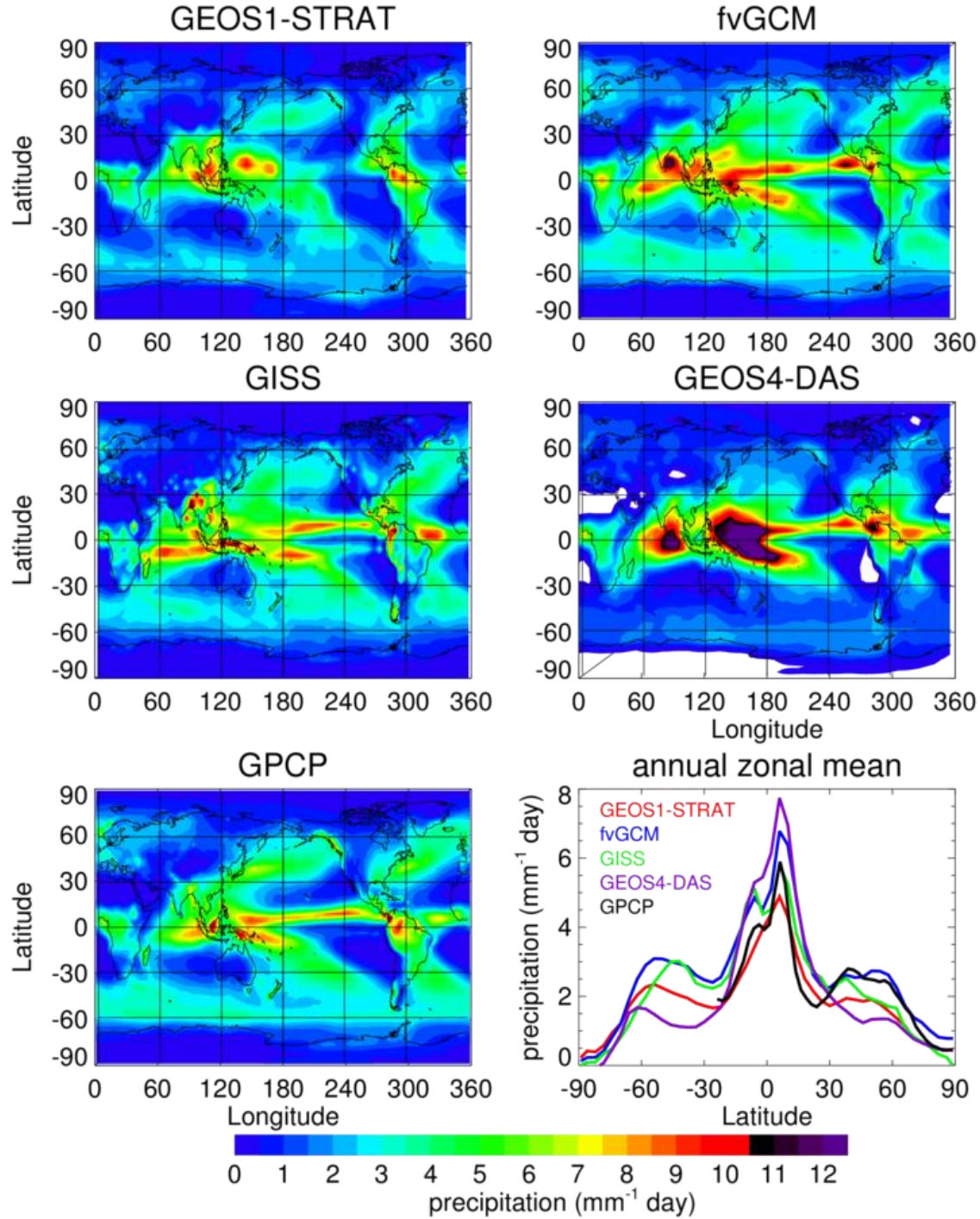
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Table 3. Annual average global budget of ^7Be in the model troposphere. The GMI model was driven by the GEOS1-STRAT, GISS II', fvGCM, and GEOS4-DAS meteorological data sets, respectively.

	GEOS1-STRAT	GISS	fvGCM	GEOS4-DAS
Burden, g	4.95 (3.86) ^b	4.00 (3.64) ^b	4.31	4.05
Residence time, days ^a	31 (29)	31 (30)	35	31
Sources, g d ⁻¹	0.22 (0.18)	0.18 (0.17)	0.18	0.19
STE	0.08 (0.04)	0.05 (0.04)	0.04	0.05
troposphere	0.14 (0.14)	0.13 (0.13)	0.14	0.14
Sinks, g d ⁻¹	0.22 (0.18)	0.18 (0.17)	0.18	0.19
dry deposition	0.01 (0.01)	0.01 (0.01)	0.01	0.02
wet deposition	0.15 (0.12)	0.12 (0.11)	0.11	0.12
radioactive decay	0.06 (0.05)	0.05 (0.05)	0.06	0.05

^aAgainst deposition only. The tropopause was determined in the model using a criterion of 2°C km⁻¹ lapse rate as defined by World Meteorological Organization. The diagnosed tropopause model layer was included as part of the troposphere.

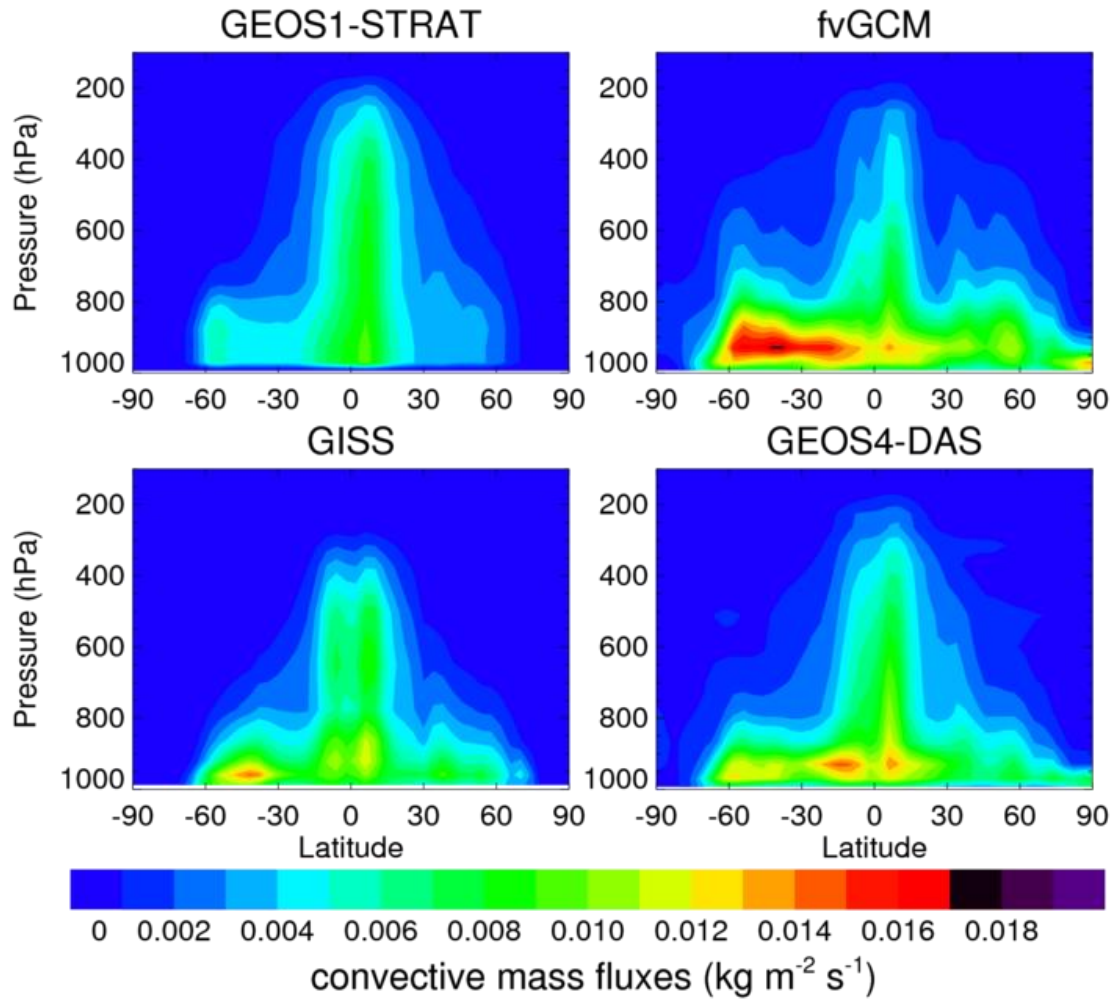
^bThe numbers in the brackets indicate the values when ^7Be cross-tropopause fluxes were adjusted for GMI/GEOS1-STRAT and GMI/GISS. See text for details.



2

3 Figure 1. Annual mean total precipitation (mm day⁻¹) at the surface in the GEOS1-STRAT, GISS
 4 II', fvGCM, and GEOS4-DAS meteorological data sets and in the observational data set from the
 5 Global Precipitation Climatology Project (GPCP, 1979-2009). Also shown is the annual zonal
 6 mean precipitation (bottom right panel).

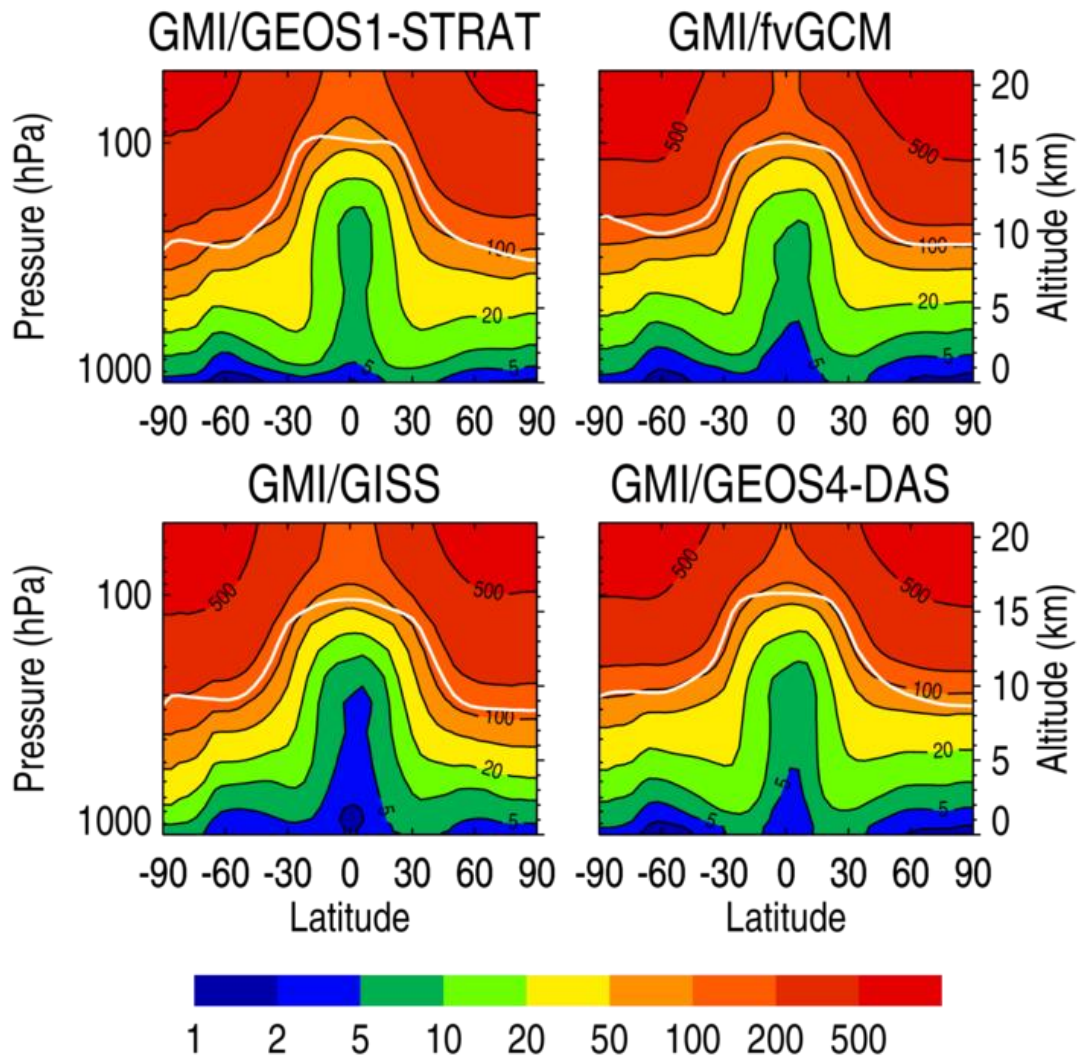
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Figure 2. Annual zonal mean convective mass fluxes ($\text{kg m}^{-2} \text{s}^{-1}$) in the GEOS1-STRAT, GISS II' GCM, fvGCM, and GEOS4-DAS meteorological data sets.

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Figure 3. Annual zonal mean mixing ratios (mBq/SCM) of ^7Be as a function of latitude and pressure (altitude), as simulated by the standard GMI CTM. The white lines indicate the annual average thermal tropopause height. Contour levels are 2, 5, 10, 20, 50, 100, 200, 500 mBq/SCM.

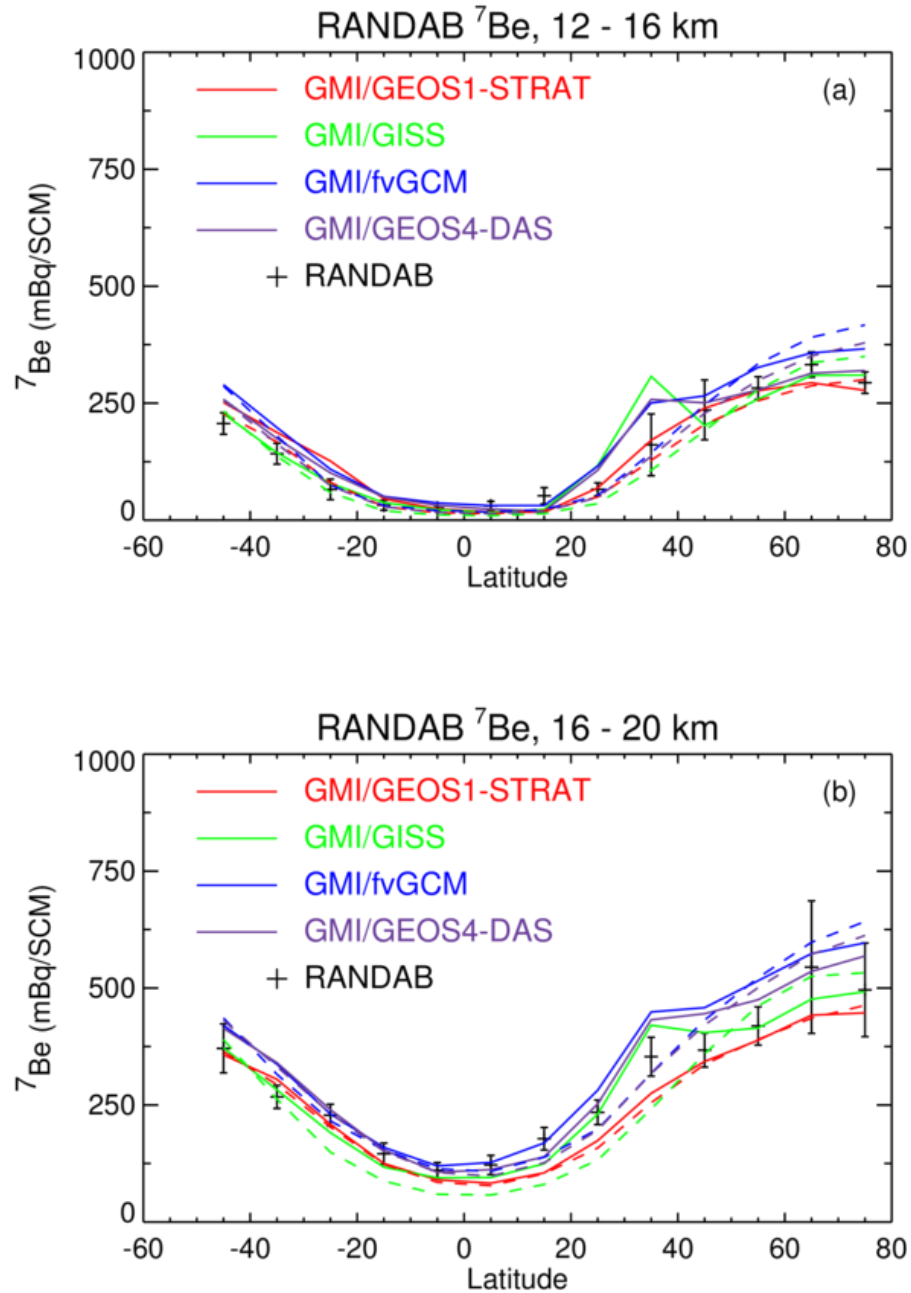


Figure 4. Observed and simulated latitudinal distributions of ^7Be in the (a) 12-16km and (b) 16-20km regions. Observed data from the EML RANDAB database are averaged into 10° bins, following Considine et al. (2005). Error bars represent ± 2 times the standard error of the averages. Model results are sampled at observation locations and month. Also shown as dashed lines are model zonal mean ^7Be concentrations to show the global representativeness of the averages constructed from sampling the simulations at the observation locations.

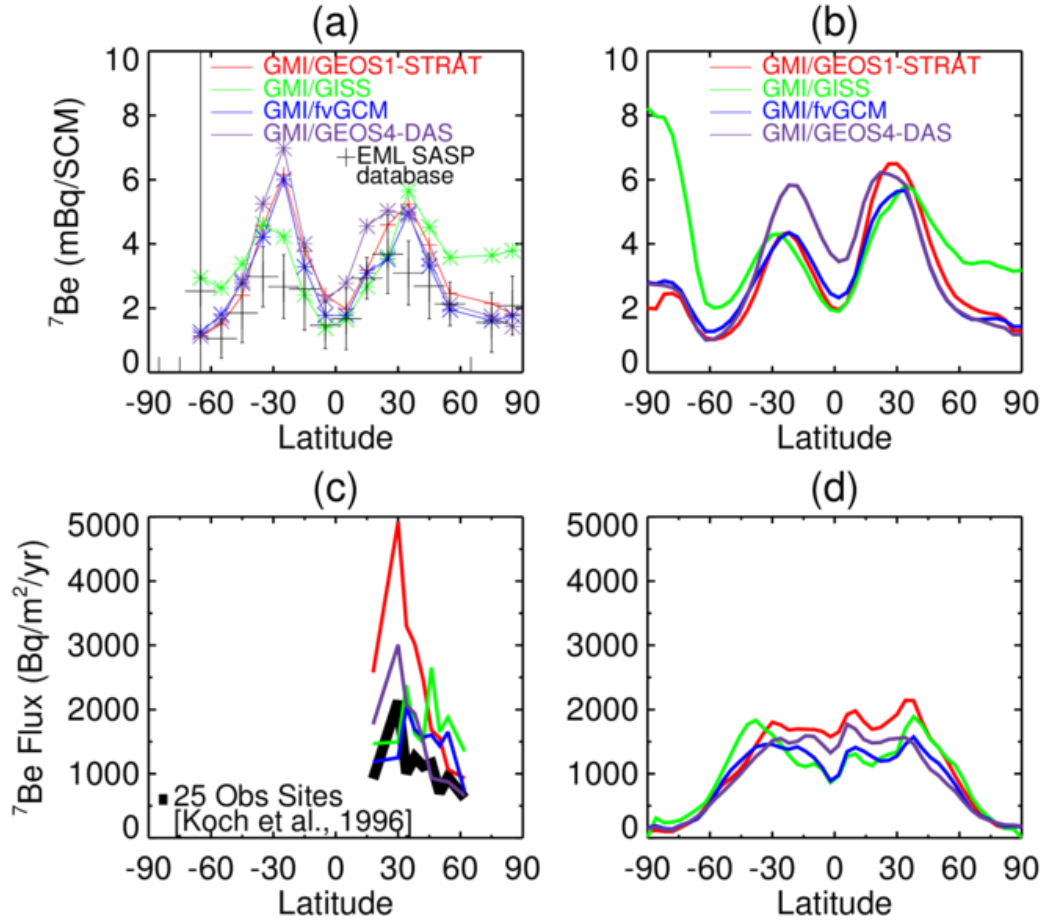


Figure 5. (a). Observed and simulated latitudinal distributions of ^7Be concentrations (mBq/SCM) near the surface. ^7Be cross-tropopause fluxes were not adjusted for the GMI/GEOS1-STRAT and GMI/GISS simulations (see Section 3). Observed data from the EML Surface Air Sampling Program (SASP) database are averaged into 10° bins. Those sites with elevation higher than 500m are not included. Error bars represent ± 2 times the standard error of the averages. Model results are sampled at observation locations and month. (b). GMI simulated annual zonal mean concentrations of ^7Be (mBq/SCM) near the surface. (c). Observed (black) and GMI simulated (color) annual mean total deposition fluxes (Bq/m²/yr) of ^7Be (at 25 sites) as a function of latitude. The data from individual sites are averaged over 4° latitude bins. The model is sampled at observation locations. (d). GMI simulated annual zonal mean total deposition fluxes (Bq/m²/yr) of ^7Be .

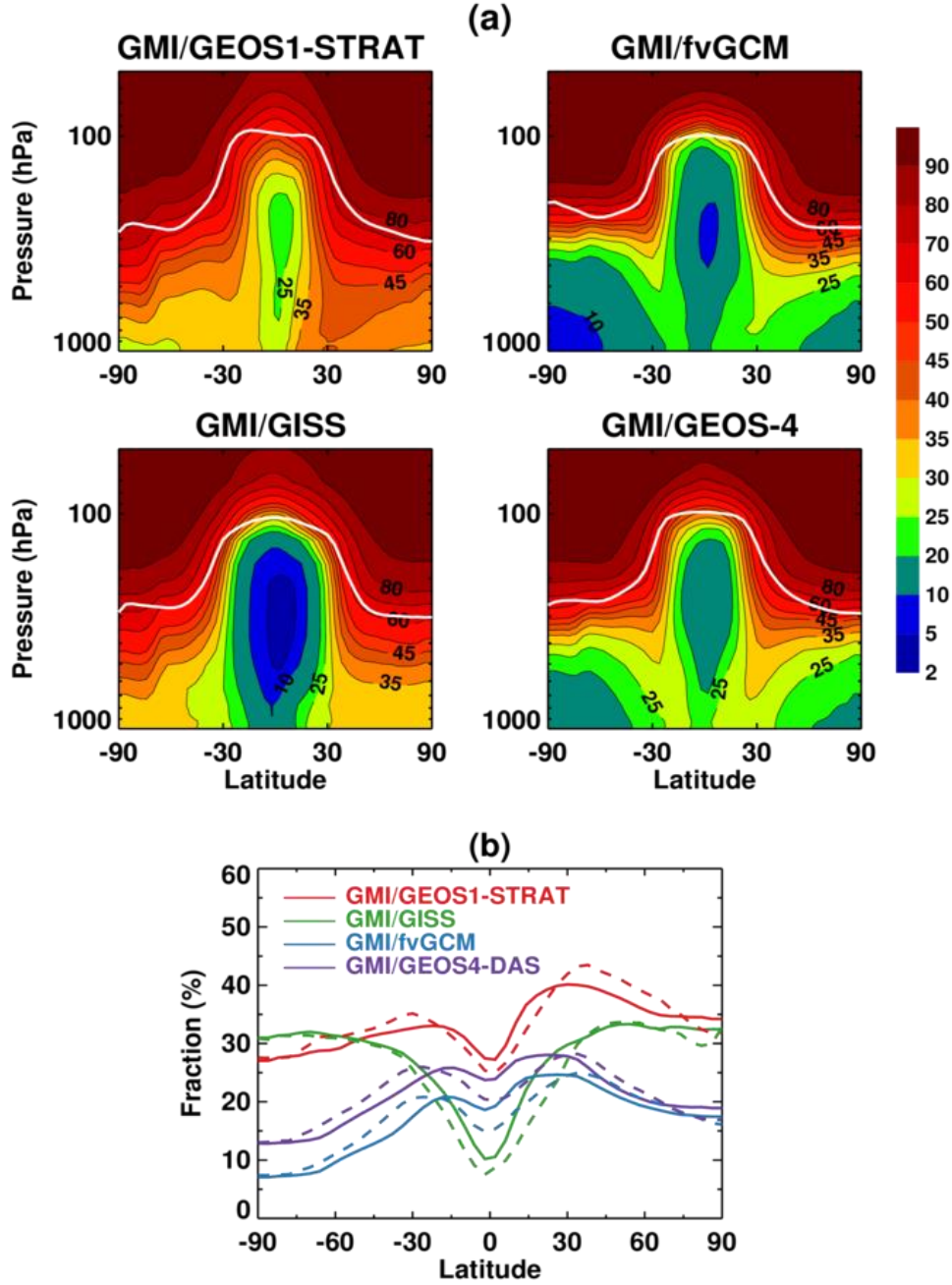
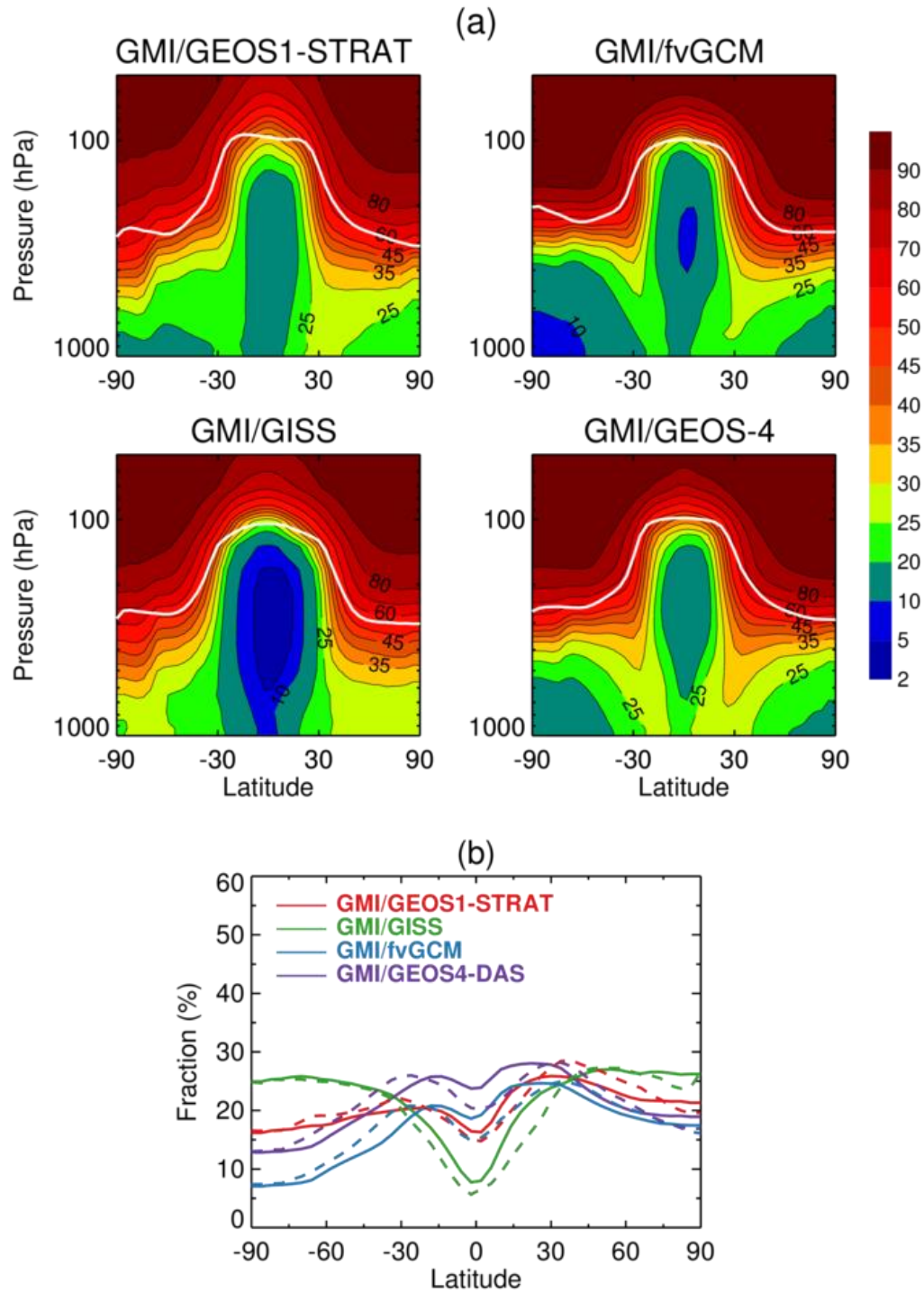


Figure 6. (a). Stratospheric fraction (%) of annual zonal mean atmospheric ^7Be concentrations in the model simulations as a function of latitude and pressure. The white lines indicate thermal tropopause height. Contour levels are 5, 10, 20, 25, 30, 35, 40, 45, 50, 60, 70, 80, 90%. (b). Stratospheric fraction of annual zonal mean surface ^7Be concentrations (solid lines) and that of annual zonal mean ^7Be total deposition fluxes (dashed lines) in the model simulations as a function of latitude.

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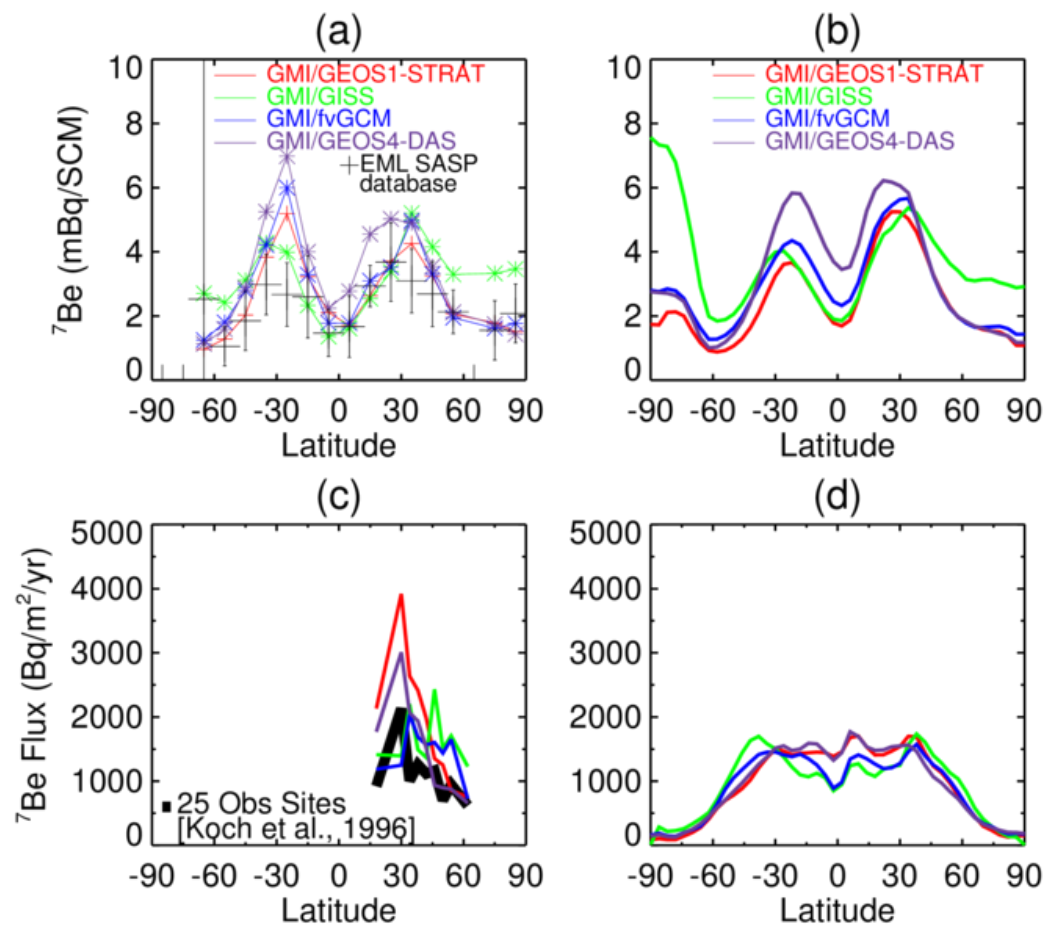


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4 Figure 7. Same as Figure 6, except that ^7Be cross-tropopause fluxes have been adjusted for
 5 GMI/GEOS1-STRAT and GMI/GISS.

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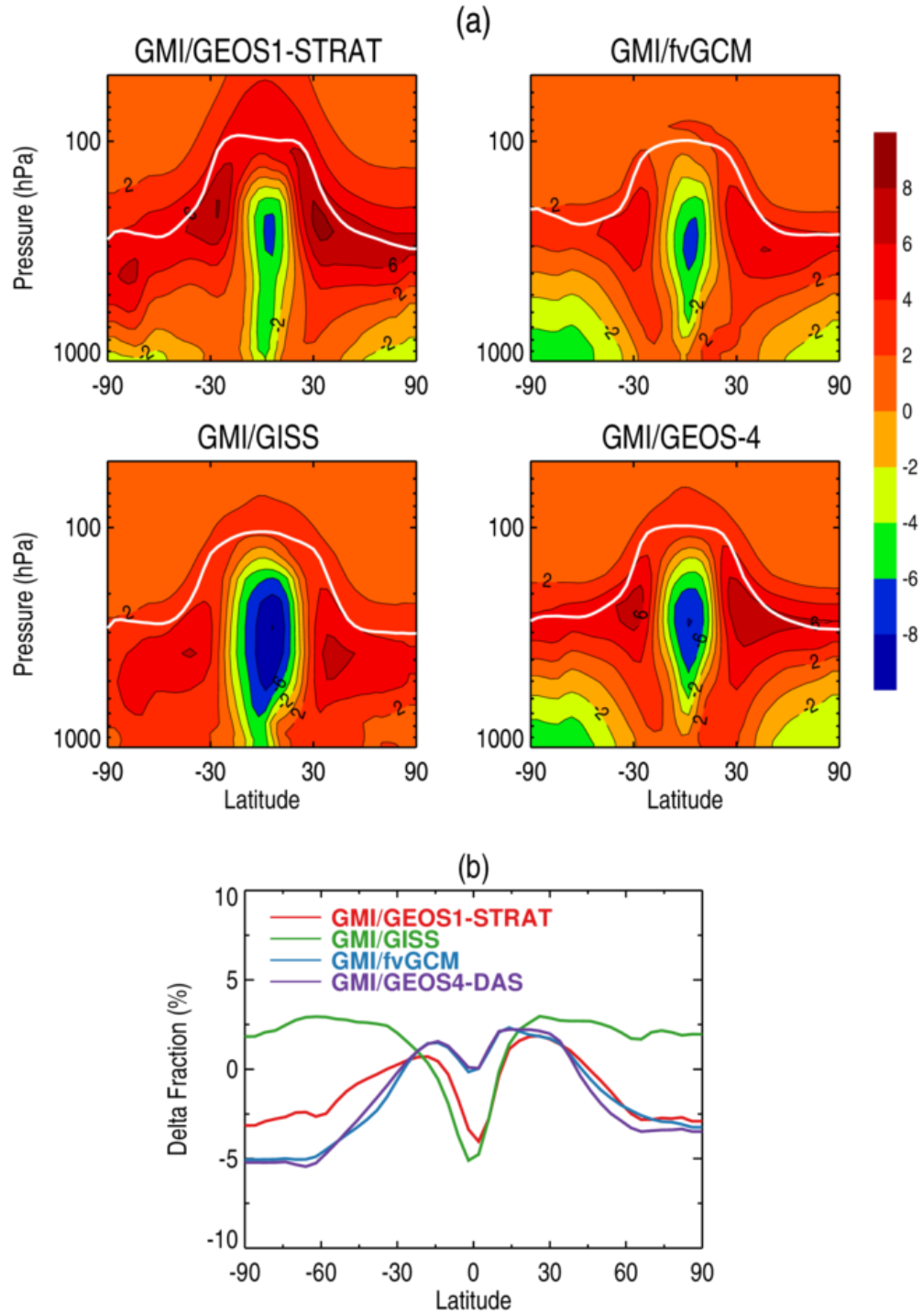


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5 Figure 8. Same as Figure 5, except that ^7Be cross-tropopause fluxes have been adjusted for
 6 GMI/GEOS1-STRAT and GMI/GISS.



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4 Figure 9. Same as Figure 6(a,b), except for the differences in the stratospheric fraction (%) of
 5 zonal mean atmospheric ^7Be concentrations between the standard simulation and a simulation
 6 where wet scavenging is turned off. Contour levels are -8, -6, -4, -2, 0, 2, 4, 6, 8%.

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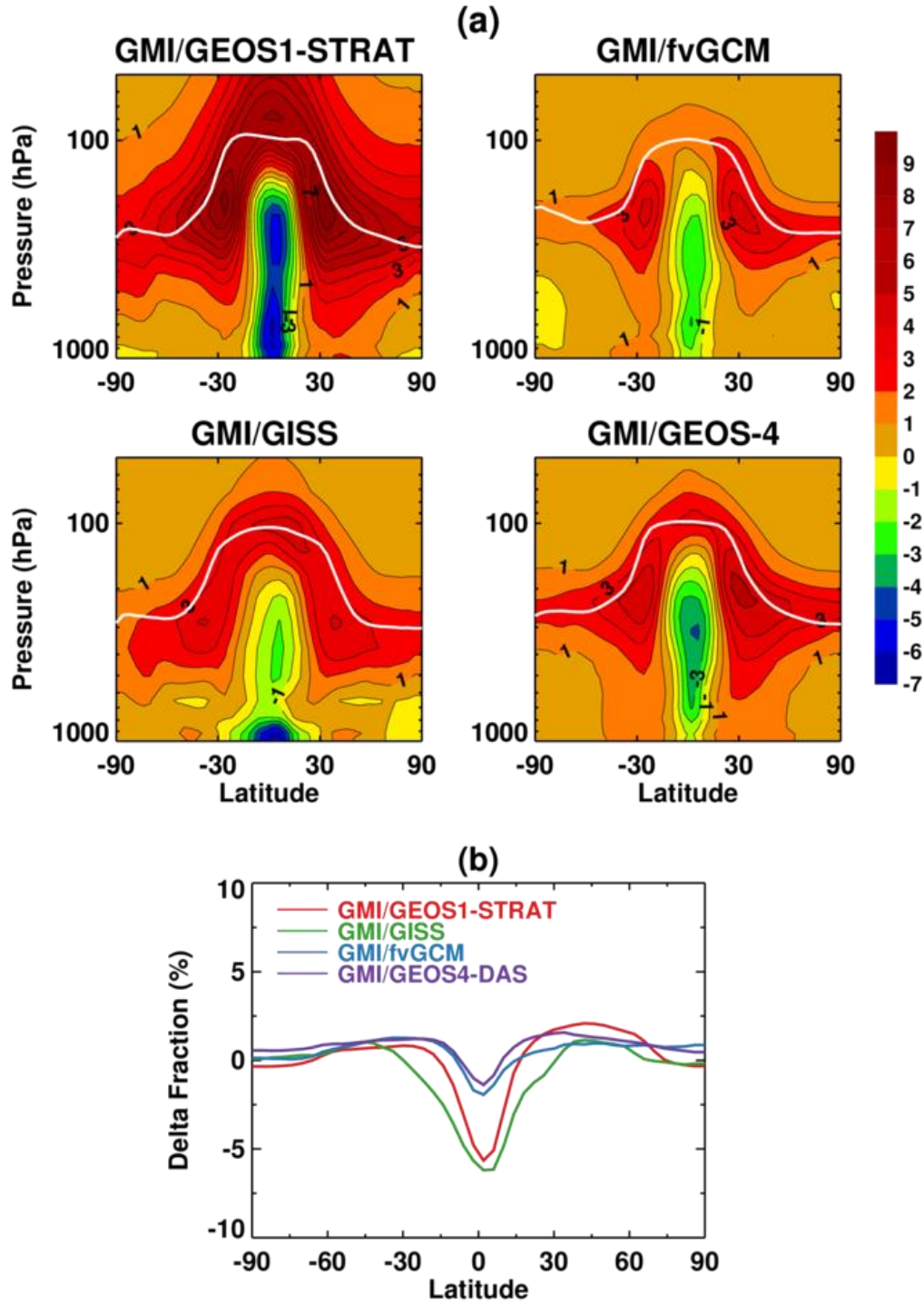
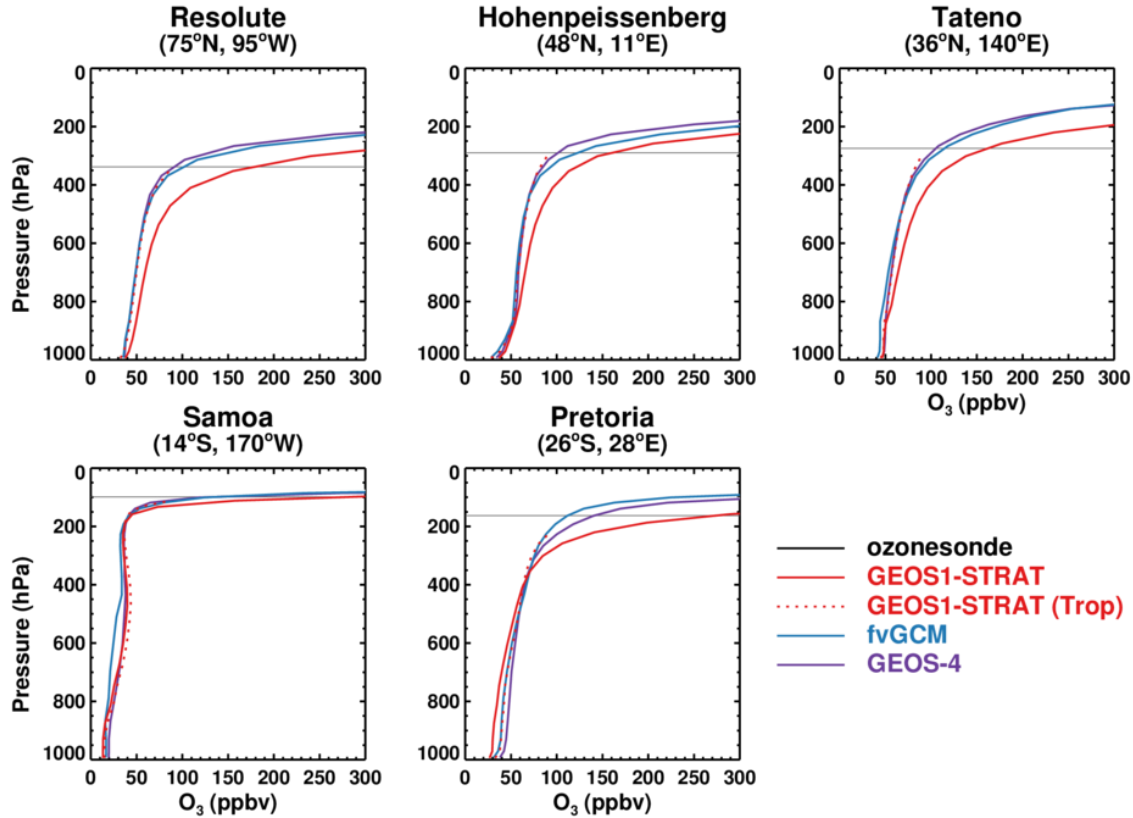


Figure 10. Same as Figure 6 (a,b), except for the differences in the stratospheric fraction (%) of zonal mean atmospheric ^7Be concentrations between the standard simulation and a simulation where convective transport and scavenging are turned off. Contour levels are -4, -3, -2, -1, 0, 1, 2, 3, 4%.



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5 Figure 11. Comparisons of GMI simulated tropospheric ozone profiles (color lines) with
 6 ozonesonde observations (black line) for a range of latitudes. Values are annual averages. Solid
 7 color lines indicate the GMI simulations. Also shown as dotted lines are tropospheric ozone
 8 profiles as simulated by the GMI tropospheric model driven by the GEOS1-STRAT
 9 meteorological field. The horizontal grey line indicates the approximate location of tropopause
 10 (i.e., the pressure level corresponding to 100 ppbv ozone concentrations in the ozonesonde
 11 observations).

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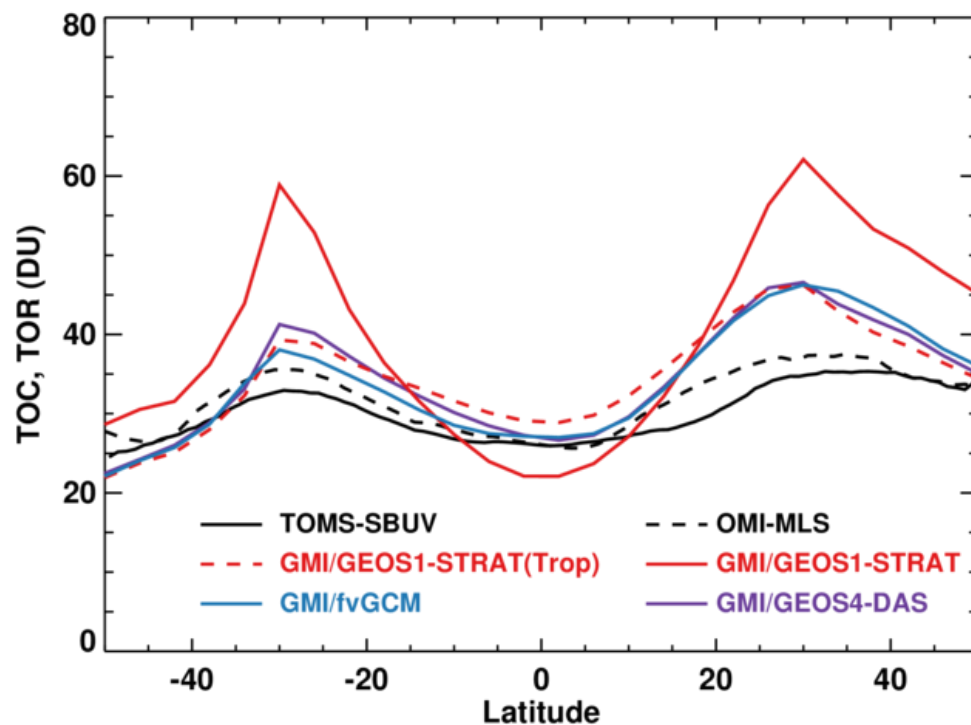


Figure 12. GMI simulated annual zonal mean tropospheric ozone column (TOC in Dobson Units) compared with observed tropospheric ozone residuals from TOMS/SBUV (1979-2005 average) and OMI/MLS (October 2004 - July 2008 average). Also shown is the annual zonal mean TOC simulated by the tropospheric version of the GMI model.

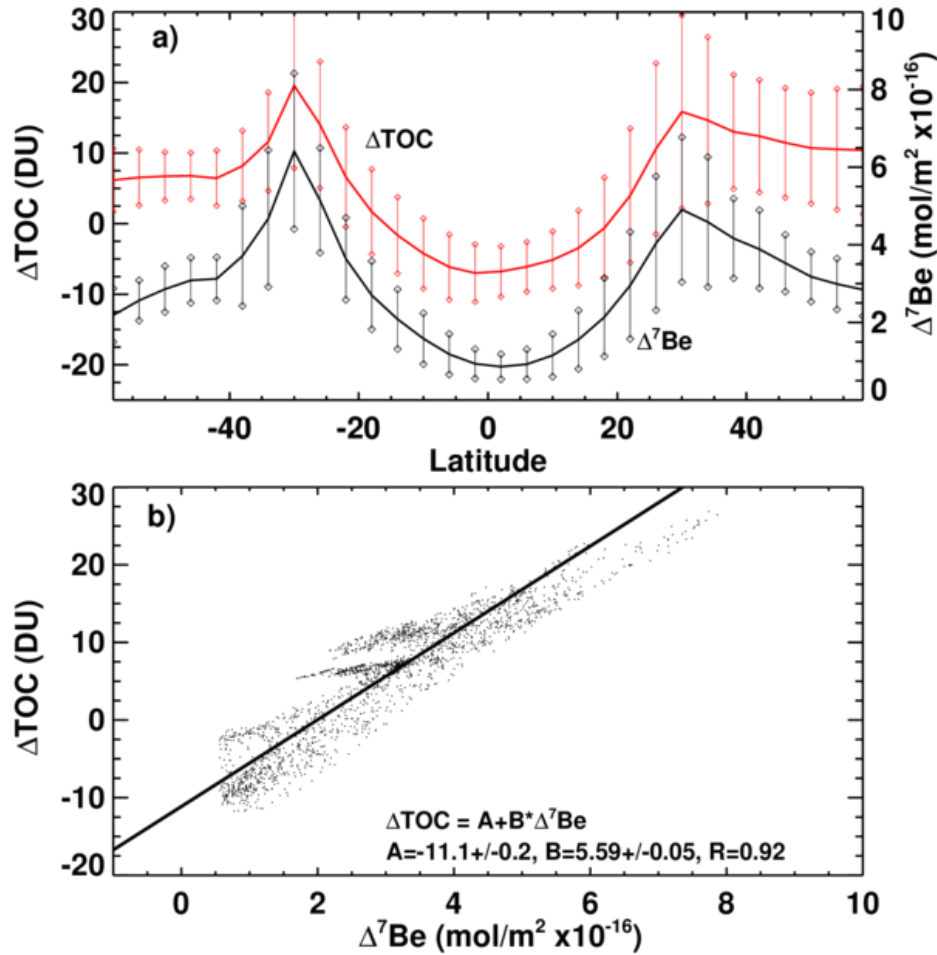


Figure 13. (a). Latitudinal variations of annual zonal mean ^7Be overestimate ($\Delta^7\text{Be}$) and tropospheric ozone column overestimate (ΔTOC) as simulated by GMI/GEOS1-STRAT. Error bars represent ± 2 times the standard error of the averages. (b). the correlation between the global distributions of $\Delta^7\text{Be}$ and ΔTOC . The lines of best fit are calculated using the reduced-major-axis (RMA) method (Hirsch and Gilroy, 1984). See text for details.