A global model of meteoric sodium

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A global model of sodium in the mesosphere and lower thermosphere has been developed within the framework of the National Center for Atmospheric Research’s Whole Atmosphere Community Climate Model (WACCM). The standard fully interactive WACCM chemistry module has been augmented with a chemistry scheme that includes nine neutral and ionized sodium species. Meteoric ablation provides the source of sodium in the model and is represented as a combination of a meteoroid input function (MIF) and a parameterized ablation model. The MIF provides the seasonally and latitudinally varying meteoric flux which is modeled taking into consideration the astronomical origins of sporadic meteors and considers variations in particle entry angle, velocity, mass, and the differential ablation of the chemical constituents. WACCM simulations show large variations in the sodium constituents over time scales from days to months. Seasonality of sodium constituents is strongly affected by variations in the MIF and transport via the mean meridional wind. In particular, the summer to winter hemisphere flow leads to the highest sodium species concentrations and loss rates occurring over the winter pole. In the Northern Hemisphere, this winter maximum can be dramatically affected by stratospheric sudden warmings. Simulations of the January 2009 major warming event show that it caused a short-term decrease in the sodium column over the polar cap that was followed by a factor of 3 increase in the following weeks. Overall, the modeled distribution of atomic sodium in WACCM agrees well with both ground-based and satellite observations. Given the strong sensitivity of the sodium layer to dynamical motions, reproducing its variability provides a stringent test of global models and should help to constrain key atmospheric variables in this poorly sampled region of the atmosphere.


1. Introduction

The morphology of the mesospheric sodium (Na) layer that originates from the ablation of meteoroids is of interest for several reasons. First, it is readily observed from the ground using lidars and provides a way to measure the state of an atmospheric region that cannot easily be measured by in situ methods. Second, the Na layer has a relatively long chemical lifetime [Xu and Smith, 2003], is advected and diffused, and therefore can be used as a tracer in the study of the dynamics of the mesosphere and lower thermosphere (MLT). Third, model studies indicate that the Na column abundances provide information on the total meteoroid mass input rate: a quantity unknown to within a factor of at least 10 [Plane, 2012]. Fourth, the Na layer interacts with the neutral and ion chemistry and therefore tests our understanding of the chemistry and energetics of the MLT. Finally, recondensation of vaporized meteoric Na “smoke” particles is thought to be a source of condensation nuclei for noctilucent clouds (NLC) [Plane, 2000]. An advantage of studying Na in the MLT is that it is observable both by ground-based instrumentation [e.g., States and Gardner, 1999; Gardner et al., 2005; Zhou et al., 2005; Xu et al., 2006; Yi et al., 2009; Yuan et al., 2012] and from satellites [e.g., Gumbel et al., 2007; Fan et al., 2007; Fussen et al., 2010]. This provides a great opportunity to validate the model dynamics and chemistry in an otherwise relatively poorly sampled region of the atmosphere.

To date, there have been few studies that have attempted to quantitatively assess the consistency between meteoroid mass input rates, their geographical and seasonal variability, and measurements of mesospheric Na density, and we know of no study that attempts to do this within a fully consistent three-dimensional chemical-dynamical model. For example, in the previous one- and two-dimensional modeling studies by Plane [2004] and Xu and Smith [2003, 2004] the ionized background atmosphere is specified from International Reference Ionosphere [Bilitza, 2003] and not calculated self-consistently with the other
model constituents. This is important since the same dynamical motions (both resolved and subgrid scale) that affect Na directly via transport will also affect it indirectly via perturbations in reaction rates caused by changes in temperature, total density, and neutral and ion mixing ratios. In the study by Gardner et al. [2005] of metal layers at the South Pole, they were unable to reproduce the observed seasonal cycle in a one-dimensional model without increasing the input of metals by a factor of 1.7 during the months of May to August. They concluded that this additional source of metals most likely came from lower latitudes, transported by the mean meridional wind. More recently, lidar observations of winds, temperature, and sodium density have revealed that they all respond to planetary wave-driven variability originating in the lower stratosphere [Yuan et al., 2012].

[4] This paper describes a new global model of Na species in the MLT that allows investigation of the effects of global-scale atmospheric motions including the mean meridional circulation, planetary waves, and atmospheric tides. The model is based on inclusion of interactive Na species chemistry and a realistic meteoritic Na source within a chemistry-climate model, both of which are described in the next section. Subsequent sections show the seasonal and short-term variability of Na species and compare them to a new, near-global, Na climatology based on observations. We also present comparisons with a number of published single-point observations. Overall, we find good agreement with observations and show that transport by the mean circulation does play a significant role in determining the seasonal cycle at high latitudes, as suggested by Gardner et al. [2005]. Our simulations indicate that temperature-dependent reaction rates and their effects on partitioning between the Na species play a secondary role compared to transport processes in determining the seasonal cycle.

2. Model Description

2.1. Whole Atmosphere Community Climate Model

[5] We utilize version 4 of the Whole Atmosphere Community Climate Model (WACCM), an extension of the Community Atmosphere Model (CAM) [Neale et al., 2013]. WACCM is a “high-top” coupled chemistry model that has an upper boundary at 5.1×10^{-6} hPa (approximately 140 km). The free-running model has been used coupled to interactive ocean and sea ice model components in the Coupled Model Intercomparison Project phase 5 [Marsh et al., 2013; Calvo et al., 2012]. Marsh et al. [2013, and references therein] describe the extensions to the model physics necessitated by including the MLT. In this study, the model horizontal winds and temperatures in the troposphere and stratosphere are constrained by reanalysis, as was done by Marsh [2011]. Comparisons of modeled upper atmosphere winds and temperatures with available climatologies are presented by Marsh [2011], Smith [2012], and Feng et al. [2013]. Running the model constrained in this way enables comparison of model predictions to observed variations over a specific period. For example, the model is able to reproduce the effects in the mesosphere of the major stratospheric sudden warming (SSW) that occurred in early 2006 [Marsh, 2011]. The reanalyses used in this study are NASA’s Modern-Era Retrospective Analysis for Research and Applications (MERRA) [Rienecker et al., 2011]. Relaxation of the model dynamical fields to those of MERRA occurs from the surface to 40 km with a time scale of approximately 10 h. Between 40 and 50 km the amount of relaxation is linearly reduced such that at 50 km the model is free-running. The model has 88 levels and an approximate vertical resolution of two grid points per scale height in the MLT. Model horizontal resolution is 1.9° in latitude by 2.5° in longitude.

[6] As in the simulations by Marsh et al. [2013], daily solar spectral irradiances used in the calculation of photolysis and shortwave heating rates are specified from the model of Lean et al. [2005] and have been adjusted to reflect the new estimate of the total solar irradiance of Kopp and Lean [2011]. The chemistry scheme is based on that of Kinnison et al. [2007], which has 59 species and 217 gas-phase chemical reactions. It includes E region ion chemistry that solves for O+, O3, N+, N2+, NO+, and electrons. Ionization occurs from particle precipitation in the auroral regions and EUV and soft X-ray photons fluxes as described by Marsh et al. [2007] and varies daily based on the observed Kp planetary geomagnetic index and the 10.7 cm solar radio flux. The model does not include modification of the ionosphere by energetic solar protons but does include the production of hydrogen and nitrogen species that would result [see, e.g., Jackman et al., 2009]. The chemistry scheme is augmented with the sodium chemistry scheme listed in Plane [2004, Table 1], which includes nine sodium species (Na, NaO, Na2O, NaOH, NaHCO3, Na+, NaCO32-, NaH2O+, and NaN22-) and 26 gas-phase chemical and 5 photolysis reactions. However, unlike in the model of Plane [2004], no assumptions are made as to whether a constituent is in photochemical equilibrium, and all species are transported. Removal of Na species from the model is via dimerization of NaHCO3, where the resulting dimer is not retained in the model:

\[ 2\text{NaHCO}_3 + M \rightarrow (\text{NaHCO}_3)_2 + M. \]  

[7] The calculated rate coefficient for the dimerization of NaHCO3 is 8.8×10^{-10} (T/200 K)^{-0.28} cm^3 molecule^{-1} s^{-1}, estimated from the dipole-dipole capture rate [Plane, 2004]. However, NaHCO3 can also polymerize with other meteoric constituent molecules, in particular FeOH. Since the relative ablation rate of Fe:Na is about 4:1 [Gardner et al., 2005; Feng et al., 2013], we increase the rate coefficient of (1) by a factor of 5. As in the recent companion paper by Feng et al. [2013], which describes mesospheric Fe chemistry in WACCM, uptake on meteoric smoke particles is treated as being equivalent to this dimerization reaction as was shown by Plane [2004]. A future version of the model with multiple meteoric metals will explicitly model the formation of meteoric dust particles from the polymerization of metal compounds and the uptake of sodium on those particles.

2.2. Meteoroid Input Function

[8] The meteoroid input function (MIF) specifies the temporal and geographical input of meteoritic metallic atoms in the MLT and is based on calculations from two recently developed models. The first model uses current knowledge of the astronomical characteristics of the sporadic meteor complex to estimate the global meteoric mass flux into the Earth’s upper atmosphere. A detailed description of this is presented by Janches et al. [2006], Fentzke and Janches [2008], and Fentzke et al. [2009].
The model estimates the meteoric mass which is deposited at each altitude within the MLT at all latitudes for each day of the year. It calculates the contribution from particles in the size range of $10^{-15}$ to $10^{-6}$ kg, which make up the bulk of metallic input into the upper atmosphere [Ceplecha et al., 1998]. Particles larger than $10^{-6}$ kg are relatively infrequent, and particles smaller than $10^{-15}$ kg will decelerate without ever reaching the high temperatures (> 1800 K) required to significantly ablate.

It is assumed that all meteoritic input originates from six main sporadic meteoroid populations, and we assign their characteristics (i.e., velocity, diurnal variability, and entry angle) accordingly, utilizing as a starting point the flux curves presented in Ceplecha et al. [1998]. Specifically, 33% of the meteors are assigned to the apex, 22% to the helion, 22% to the antihelion, 11.5% to the north toroidal, and 11.5% to the south toroidal. This enables calculation of the time evolution of the population's incoming angular and velocity characteristics in a given geographical location.

The model has been validated against High Power and Large Aperture radar head echo observations [Janches et al., 2006; Fentzke and Janches, 2008; Fentzke et al., 2009; Pifko et al., 2013]. Unlike those studies, however, we consider all particles large and fast enough to ablate, without considering if the amount of electrons produced during entry is large enough for detection by a sensitive radar [Janches et al., 2008]. The result is the distribution of particles as a function of time, initial mass, initial absolute velocity, and entry angle accordingly, utilizing as a starting point the flux curves presented in Ceplecha et al. [1998]. Specifically, 33% of the meteors are assigned to the apex, 22% to the helion, 22% to the antihelion, 11.5% to the north toroidal, and 11.5% to the south toroidal. This enables calculation of the time evolution of the population's incoming angular and velocity characteristics in a given geographical location.

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3. Results: Global Distribution and Seasonal Variations

Finally, we integrate the ablation profiles over the distribution of incoming particles to obtain a Na input profile as a function of day of the year, latitude, and height. The annual cycle of the column integral of the Na input used in WACCM simulations is shown in Figure 1. The same cycle is used for every year of simulation. In the extratropics the cycle is annual, peaking in September in the Northern Hemisphere (NH) and March in the Southern Hemisphere (SH) with input rates in excess of 2400 atoms cm$^{-2}$ s$^{-1}$. The amplitude of the cycle is largest near the poles, where it reaches about 28% of the annual mean. In the tropics the MIF is relatively constant at around 2000 atoms cm$^{-2}$ s$^{-1}$. The annual and global mean input of sodium is $1.75 \times 10^8$ atoms cm$^{-2}$ d$^{-1}$. Integrated over the globe, the sodium mass input is 0.035 t d$^{-1}$. Assuming a relative elemental abundance of 0.8% [Vondrak et al., 2008], the total ablated meteoritic mass input would be equivalent to 4.6 t d$^{-1}$. We note that this is at the low end of the range of the estimates of the total (ablated and non-ablated) daily input of cosmic dust input which extends to nearly 300 t d$^{-1}$ [Plane, 2012]. Possible changes to the model that could permit a higher mass input are considered in section 5 and by Feng et al. [2013].
In Figure 3 the latitudinal and seasonal variations of the monthly mean column integrals of Na⁺, Na, and NaHCO₃ are shown. Extratropical Na⁺ peaks in late summer and of the three species resembles the MIF variations the most. In contrast, extratropical Na and NaHCO₃ have maxima in winter. The largest Na column densities are seen in the high-latitude Southern Hemisphere during June and July, where the total column exceeds $9 \times 10^{9}$ atom cm⁻². In the tropics Na⁺ and NaHCO₃ are relatively constant, whereas Na has a semiannual variation with maximum values during equinoct. Inspection of the sum of the three species (Figure 3d) reveals that the drastically different seasonal variations cannot simply be caused by a chemical repartitioning of the available meteoritic Na species family, since the sum would then also follow the MIF. Instead, the model predicts that the largest total column values occur over the SH winter pole, again indicating that convergence of the Na flux over the poles plays a role.

Figure 4 shows the ratio of Na⁺ to Na calculated from the monthly means shown in Figure 3. The ratio ranges from below 0.4 to above 3. Averaged over all seasons and latitudes, the ratio is close to unity. This is larger than the ratio determined from rocket-borne mass spectrometric Na⁺ measurements of between 0.2 and 0.25 by Plane [2004]. However, that estimate is based on just five rocket flights [Kopp, 1997] that did not simultaneously measure Na and do not represent an annual and global average (all flights lie between 37 and 68°N). Also, the rocket profiles reported by Kopp [1997] show very large variability in their vertical structure, as might be expected from a single-point measurement. Still, if we average the modeled ratio at the latitude and month of the rocket flights, we obtain a ratio 0.9, which, while on the lower end of the modeled range of ratios, is larger than derived from observations. A similar discrepancy was reported by Plane [2004], who states that the modeled ratio could be reduced by reducing the average entry velocity of incoming meteors such that they ablate lower in the atmosphere. Work is ongoing to evaluate if such a reduction is compatible with the astronomical model of meteor fluxes described in section 2.2.

The seasonal variation of the altitude of the peak of the Na layer is shown in Figure 5. The altitude was calculated by finding the geometric height of the maximum of a quadratic polynomial fit to the sodium density at the three levels that span the peak sodium density. In choosing three points, the fit is unique and easily differentiated to find the height of the maximum. At middle to high latitudes the seasonal cycle is predominantly annual, with the peak of the layer occurring at a higher altitude during summertime, likely caused by upwelling over the summer pole. In the

Figure 2. Global mean distribution of sodium compounds during July.

Figure 3. Monthly mean total column ($10^{9}$ cm⁻²) of (a) Na⁺, (b) Na, (c) NaHCO₃, and (d) their sum.
Figure 4. Ratio of Na⁺ to Na monthly mean column shown in Figures 3a and 3b. Contour levels are [0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, 1, 1.2, 1.5, 2, 3, 4, 5].

tropics, there is a semiannual oscillation in the layer peak height, with minima in April and September/October. The annual mean value varies with latitude between 89.8 and 87.5 km, with the tropics higher than the high latitudes. The global annual mean layer peak height is 89.1 km.

Figure 6 shows the latitude and time variation of zonal mean column sodium from years 2006 to 2009. Differences between years are the result of internally generated variability, variations in external forcing (e.g., solar variability), and the MERRA reanalysis. Overall, it appears that the seasonal cycle is consistent from year to year. There does appear to be a large amount of day-to-day variability over the winter poles. In particular there is the very large increase above 60°N in February of 2009. This is preceded by lower column abundances in late January. To better visualize this rapid change, Figure 7 shows the NH polar cap on 22 January and 6 February 2009. On 22 January the column abundance is below 6 x 10^9 atom cm⁻² almost everywhere. In contrast, on 6 February they exceed 10^10 atom cm⁻² over most of the pole northward of 60°N and can reach 2 x 10^10 atom cm⁻² above 80°N. The almost quadrupling of the Na column in 2 weeks is most likely associated with the major stratospheric sudden warming (SSW) that occurred on 24 January 2009 [Butler and Polvani, 2011; Yuan et al., 2012].

To illustrate this further, the NH polar cap temperature variations in the stratosphere and lower thermosphere during the first 100 days of 2008 and 2009 are shown in Figure 8. Between day 12 and day 23 of 2009, temperatures at 6.9 hPa increased by over 60 K, and at 0.002 hPa there was a major cooling. Over the following 40 days the stratospheric temperature slowly returned to its prewarming level. In the lower thermosphere, temperatures rebounded much more quickly and exceeded the prewarming values by almost 20 K. Temperatures stayed elevated in the lower thermosphere for several weeks following the rebound. In contrast, 2008 temperature variations were more sporadic, with many short-lived temperature swings. As discussed by Marsh [2011], the dramatic cooling and later heating in the lower thermosphere modeled in WACCM is caused by adiabatic heating from anomalous vertical motions over the pole. The changes in the vertical wind are the result of changes in the mean meridional wind, which in turn is caused by changes in gravity wave momentum deposition. Gravity wave propagation through the lower atmosphere is modified as the stratospheric mean winds change during the major warming. The result is that during the SSW the mean meridional wind reverses direction and flows away from the pole and leads to anomalous upwelling and cooling. Following the SSW, the wind returns to the normal poleward and downward flow, but it is stronger than prior to the SSW, and hence a strong mesospheric warming is simulated.

One possible driver for the increase of Na by a factor of 3 (shown in Figure 8c) following the SSW then is via the effects of temperature changes in reaction rates. For example, the temperature dependence of neutral Na chemistry is mostly governed by the following reaction [Plane, 2004]:

\[ \text{NaHCO}_3 + \text{H} \rightarrow \text{Na}^+ + \text{H}_2 + \text{CO}_2, \quad k = 1.8 \times 10^{-13} T^{0.777} \exp(-1014/T), \]

An increase of temperature would have the effect of increasing the net conversion of NaHCO3 to Na. Increased downwelling of atomic H might also be important in converting NaHCO3 to Na. However, conversion of the major reservoir species NaHCO3 to Na is unlikely to explain the sudden rise, since this species increases following the SSW (see, e.g., Figure 8c). We note that Na⁺ shows a similar temporal variation to NaHCO3 and also increases following the SSW (not shown).

Since a repartitioning of available Na species within the polar cap is therefore unlikely to explain the rapid decrease and increase, it leaves transport in and out of the cap as the cause. The upwelling and equatorward flow during the SSW would bring air poor in Na from lower altitudes and distribute this over the polar cap. Then, following the SSW, the enhanced downward and poleward flow would tend to transport Na species from midlatitudes into the polar region.
This is clearly seen in Figure 8d), which shows the column-integrated flux of Na species across the 70°N latitude circle divided by the area of the polar cap. We find that following the SSW the flux into the cap would be sufficient to increase the average column of Na species by almost $3 \times 10^9$ cm$^{-2}$ per day. The poleward and downward flow would not only increase all Na species but also promote conversion of Na$^+$ to Na due to enhanced recombination rates as the density increases with downward motion. A similar mechanism is likely the cause of the low Na$^+$ to Na ratios seen in Figure 4 over the wintertime pole.

4. Comparison With Observations

4.1. A Na Reference Atmosphere

[21] In order to compare the WACCM model results with observations, we first constructed a reference atmosphere. This consists of zonally averaged data in 10° latitude bins on a monthly time scale. The reference atmosphere draws on several sources. The most comprehensive is the recent near-global set of satellite limb-scanning measurements made with the Optical Spectrograph and Infra-Red Imager System (OSIRIS) spectrometer on the Odin satellite [Fan et al., 2007]. Because the data are self-consistent and ground-truthed against lidar observations [Gumbel et al., 2007], it forms the backbone of the reference atmosphere. However, there are some limitations. First, the data cover only two complete years, 2003 and 2004. Second, the sun-synchronous orbit of Odin provided measurements only at about 0600 and 1800 local time. Since the Na layer is subject to photochemical and tidally driven diurnal variations [Clemesha et al., 1982; Fan et al., 2007], the data from both local times have been averaged. Third, OSIRIS measures Na resonance fluorescence in the dayglow, so measurements are restricted to periods when the mesosphere is illuminated (solar zenith angle $<92^\circ$), and so there are no data at midlatitudes to high latitudes during winter. In order to partly overcome these limitations, lidar data from the South Pole for the years 1995–1997 [Gardner et al., 2005], from São José dos Campos (23°S) for the years 1972–1986 [Clemesha et al., 2004], and Urbana-Champaign (40°N) and Fort Collins (41°N) for the years 1991–1999 [Plane et al., 1999; States and Gardner, 1999; She et al., 2000] have been included. Lidar data from other observatories are either
already covered by the satellite measurements or have not yet been published as a long-term record. It should be noted that the uncertainty in the absolute Na density near the layer peak retrieved from OSIRIS is ±10% [Gumbel et al., 2007], similar to modern ground-based lidars. However, the natural variability of the Na layer means that the average monthly column abundance and peak density values in some of the 10° latitude bins have a larger uncertainty (up to ±30%), depending on the number of profiles in the average [Fan et al., 2007]. The global map of Na column abundance was constructed starting from the satellite measurements in Fan et al. [2007, Figure 1]. This is a grid map of zonal monthly averages in 10° latitude bins. Available monthly averaged lidar data (see above) were then added to grid boxes where there were no OSIRIS data. The remaining empty grid boxes were then filled in by interpolation. The largest uncertainty in this exercise is the region between 30 and 80°S, between April and August, where there are no measurements available. The Na column abundance at high northern latitudes in winter is consistent with the short lidar record published by Tilgner and von Zahn [1988].

Although these data sets are taken from different decades and phases of the solar cycle, a long-term study of the Na layer [Clemesha et al., 2004] shows that, at least at low latitudes (23°S), the effects of changing climate and the solar cycle on the Na layer are small. The centroid of the layer moves down only 0.17 ± 0.11 km between solar minimum and maximum. This downward trend is likely due...
to changes in chemistry: at solar maximum there is a greater plasma density in the lower $E$ region and so more Na is converted to Na$^+$ on the topside, whereas higher atomic H and O concentrations below 90 km (produced by increased Lyman-$\alpha$ radiation) recycle Na from NaHCO$_3$ and other reservoir species more rapidly on the underside of the layer. Measurements made at Urbana-Champaign, Illinois ($40^\circ$N) in 1991–1994 [Plane et al., 1999] and 1996–1998 [States and Gardner, 1999], which correspond to periods at and shortly after solar maximum and minimum, respectively, show that the Na column abundance was about 20% higher at solar maximum.

The resulting Na column abundance, as a function of latitude and month, is shown in Figure 9a. The average column abundance is $3.9 \times 10^{9}$ atom cm$^{-2}$, although this ranges by a factor of 20 (0.33–7.4 $\times 10^{9}$ atom cm$^{-2}$), depending on time and location. Abundances exhibit a seasonal variation very similar to WACCM with a wintertime maximum in the extratropics: October–November in the NH, and June–July in the SH. As in the model, the size of the variation is latitude dependent: at low latitudes the winter enhancement is only a factor of about 1.3, whereas at midlatitudes the wintertime enhancement is approximately a factor of 3 and more than 10 in the polar regions. In the tropics the variation is semiannual, peaking at equinox. The shift from an equatorial semiannual to polar annual cycle in both the observational climatology and WACCM confirms the finding of Fussen et al. [2010] based on observations from the Global Ozone Monitoring by Occultation of Stars (GOMOS) satellite.

The agreement in the height of the layer (shown in Figure 9b) is not as good, with observed heights about 2–3 km higher than the model. The exact cause of this offset is difficult to assess, although it is within a model vertical grid spacing. The discrepancy may be related to errors in the background atmosphere density profile versus geometric altitude, which depends on the thermal structure. As noted by Smith [2012] the mesopause temperature in WACCM is warmer than satellite observations made by the Sounding the Atmosphere by Broadband Emission Radiometry instrument by over 10 K throughout the tropics and subtropics. However, the model does reproduce much of the observed latitudinal structure, with the layer being overall higher in the tropics and higher during summertime in the extratropics.

### 4.2. Lidar Comparisons

The modeled and observed annual cycles of monthly mean column sodium at Fort Collins ($41^\circ$N, $104^\circ$W) and the South Pole are shown in Figure 10. Both observational estimates are from lidar measurements: Fort Collins data are listed in She et al. [2000, Table 2], and the South Pole data are taken from the study by Gardner et al. [2005, Table 1]. Also shown are the zonal mean seasonal cycles from Figure 9a and the empirical fit to GOMOS measurements [Fussen et al., 2010]. Both model and observations show a predominantly annual cycle, with minimum during the summer, consistent with Figure 3.
[26] The amplitude of the seasonal cycle is larger at the South Pole location compared to Fort Collins. WACCM is in better agreement with observations at Fort Collins than South Pole, with model values larger than observed during February, March, and July. The values from the GOMOS model are generally lower at the South Pole than the other estimates in seasons other than summer, but since GOMOS did not make observations at the South Pole during this time [Fussen et al., 2010], the values there are likely an extrapolation from lower latitudes where the seasonal cycle is weaker. The summer minimum is significantly lower at the South Pole than at Fort Collins. A similar difference between observations at the South Pole and Urbana Atmospheric Observatory (40°N) was seen by Gardner et al. [2005]. They attributed the very low values at the South Pole to the uptake of sodium by NLC particles. Since this process is not included in these WACCM simulations, the very low sodium abundances that are seen in December and January are related more to the effects of dynamics. We note that removal of Na species via dimerization is a second-order reaction with a rate proportional to the square of the Na species density, whereas removal on NLC particles is proportional to the product of the Na species density and the NLC volumetric surface area, making dimerization more efficient where densities are large. The Na layer peak is at its highest altitude in summer, further distancing it from the maximum in the NLC volumetric surface area, which is around 87 km [Plane et al., 2004]. Over the summer pole, densities of Na and NaHCO3 at 87 km are around 10 and 1 cm⁻³, respectively, so that even if uptake on NLC is very efficient it would not change the seasonal cycle of the column significantly. It should be noted that metals that have their maximum abundance lower in the atmosphere, such as Fe, will be more affected by NLC uptake, and this has been included in the model of Feng et al. [2013].

5. Discussion and Conclusions

[27] The relative importance of chemistry and transport in determining the seasonal cycle depends on the lifetimes for these processes. Considering all Na-bearing species, the MIF is the only source, and dimerization is the only loss. If the chemical lifetime (i.e., the inverse of the loss rate) is short compared to that of transport, then the distribution of Na species and their loss should look very much like the MIF. The latitudinal distribution, averaged over the years 2005 and 2008, of the column-integrated MIF and the zonal mean Na species removal via (1) are shown in Figure 11a. The MIF is relatively constant with respect to latitude, whereas the losses are greatly enhanced in the polar regions and lower than the meteoritic input at latitudes equatorward of 40°. Over the long-term, mass continuity dictates that the time average of differences between chemical production and loss (Δ) are balanced by transport processes. Since Figure 11 shows annual and zonal averages of the column-integrated production and loss, that transport process must be related to net meridional transport. This can be expressed mathematically as follows:

\[
\frac{1}{\rho S} \frac{\partial (\rho S \tilde{\chi})}{\partial t} = \left\{ \frac{1}{\alpha \cos \phi} \frac{\partial}{\partial \phi} \left( \rho S \tilde{\chi} \cos \phi \right) \right\},
\]

where \( \tilde{\chi} \) is zonal mean mixing ratio, \( \rho \) is density, \( \alpha \) is latitude, \( \alpha \) is the radius of the Earth, and \( v^* \) is the transformed Eulerian mean meridional wind [Smith et al., 2011]. Angle brackets denote the vertical integral over all altitudes. The last term on the right-hand side is the divergence of the Na species mean meridional transport (dashed line in Figure 11b). It is clear that much of the imbalance between production and loss is related to meridional transport. We see clearly that transport serves to move Na species away from the equator toward the poles where it is lost.

[28] Overall, the model appears to be capable of reproducing the seasonal and short-term variability of meteoric Na. This is achieved with a MIF model that has a total meteoric mass input of 4.6 t d⁻¹, mostly contributed by meteoroids with masses between 10⁻¹⁵ and 10⁻⁹ kg. Even though this amount is within the lower limit of estimates of this hotly debated quantity derived from a very diverse pool of observational techniques and measurements [Plane, 2012], it appears to be sufficient to model the global Na layer with very little or no scaling. We note, however, that there are several parameters in WACCM that could be adjusted to allow for a larger meteoric input, for example, the ad hoc increase in the dimerization of NaHCO₃ to account for the absence of other meteoritic metals. Future versions of the models will include additional metal species to better represent this loss process. Coupling WACCM to a sectional microphysics code that models the growth of meteoric dust such as described by Bardeen et al. [2008] would also permit uptake of metals on these particles.

[29] In addition, the eddy diffusion coefficient can be increased by decreasing the effective Prandtl number [Smith et al., 2011], which would lead to downward transport of Na and quicker conversion to NaHCO₃. Sensitivity tests in WACCM with Fe chemistry show that halving the Prandtl...
number (effectively doubling the eddy diffusion) could decrease Fe densities near the layer peak by up to a third [Feng et al., 2013]. However, there are limits on adjusting the Prandtl number, since it also is used in the calculation of diffusion of heat and other trace species. Also, drastically increasing the loss rate will diminish the importance of transport, as discussed above, and reduce the magnitude of the high-latitude seasonal cycle. If the dimerization rate coefficient is increased to make irreversible chemi-

References


