

**Simulation and observations of stratospheric aerosols
from the 2009 Sarychev volcanic eruption**

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KRAVITZ ET AL.: SARYCHEV OPTICAL DEPTH

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87 **Abstract**

88 We used a general circulation model of Earth's climate to conduct simulations of the 12-
89 16 June 2009 eruption of Sarychev volcano (48.1°N, 153.2°E). The model simulates the
90 formation and transport of the stratospheric sulfate aerosol cloud from the eruption and the
91 resulting climate response. We compared optical depth results from these simulations with limb
92 scatter measurements from the Optical Spectrograph and InfraRed Imaging System (OSIRIS), in
93 situ measurements from balloon-borne instruments lofted from Laramie, Wyoming (41.3°N,
94 105.7°W), and five lidar stations located throughout the Northern Hemisphere. The aerosol
95 cloud covered most of the Northern Hemisphere, extending slightly into the tropics, with peak
96 backscatter measured between 12 and 16 km in altitude. Aerosol concentrations returned to near
97 background levels by Spring, 2010. After accounting for expected sources of discrepancy
98 between each of the data sources, the magnitudes and spatial distributions of aerosol optical
99 depth due to the eruption largely agree. In conducting the simulations, we likely overestimated
100 both particle size and the amount of SO₂ injected into the stratosphere, resulting in modeled
101 optical depth values that were a factor of 2-4 too high. Model results of optical depth due to the
102 eruption show a peak too late in high latitudes and too early in low latitudes, suggesting a
103 problem with stratospheric circulation in the model. The model also shows a higher annual
104 decay rate in optical depth than is observed, showing an inaccuracy in seasonal deposition rates.
105 The modeled deposition rate of sulfate aerosols from the Sarychev eruption is higher than the
106 rate calculated for aerosols from the 1991 eruption of Mt. Pinatubo.

107 **1. Introduction**

108 Sarychev Volcano (48.1°N, 153.2°E) in the Kuril Islands, Russia, erupted (Figure 1) over
109 the period 12-16 June 2009, injecting approximately 1.2 Tg of sulfur dioxide into the lower
110 stratosphere at an altitude of approximately 11-16 km [Haywood *et al.*, 2010]. This was the
111 second major stratospheric injection of SO₂ in the span of a year, the previous one being the
112 eruption of Kasatochi on 8 August 2008 [Kravitz and Robock, 2010]. The largest eruptions prior
113 to these were Mount Pinatubo and Mount Hudson in 1991 [Carn and Krueger, 2004].

114 The climate effects of volcanic eruptions are well established [Robock, 2000]. These
115 effects are due to the production of a large layer of sulfate aerosols in the stratosphere, which
116 efficiently backscatters solar radiation, effectively increasing the planetary albedo and causing
117 cooling at the surface. For these radiative effects to accumulate, the aerosols must remain in the
118 atmosphere for an extended period of time. Stratospheric volcanic aerosols have an average *e*-
119 folding lifetime of 1 year [Budyko, 1977; Stenchikov *et al.*, 1998; Gao *et al.*, 2007]. Were the
120 injection to occur only into the troposphere, the climate effects would be greatly muted, as the
121 atmospheric lifetime of tropospheric aerosols is about a week [Seinfeld and Pandis, 2006].

122 Determining the climate effects requires an accurate assessment of the amount of sulfate
123 aerosols created in the stratosphere, as well as the spatial and temporal patterns of the aerosol
124 layer. General circulation models are useful predictive tools for estimating volcanic effects, and
125 they have been used with great success in replicating the effects of past volcanic eruptions [e.g.,
126 Oman *et al.*, 2006a]. However, any model can benefit from further testing and improvement. As
127 such, we use the recent eruption of Sarychev to test one climate model's ability to accurately
128 create and transport sulfate aerosols.

129 *Kravitz et al.* [2010] compared modeled results of sulfate aerosol optical depth with
130 satellite and ground-based retrievals from the Kasatochi eruption. Although the spatial pattern of
131 aerosol distributions in the model and the observations largely agreed, they discovered a
132 discrepancy of an order of magnitude in the actual values. They were able to explain some of
133 this discrepancy, but a factor of 2-4 remained unexplained. A similar comparison between
134 model results and observations of the eruption of Sarychev will allow us to expand this study and
135 better analyze the discrepancy. As in *Kravitz et al.* [2010], a large part of our comparison will be
136 with the Optical Spectrograph and InfraRed Imaging System (OSIRIS), a Canadian instrument
137 on the Swedish Odin satellite [*Llewellyn et al.*, 2004]. Launched in 2001 and still currently
138 operational, OSIRIS measures the vertical profile of limb-scattered sunlight spectra. Previous
139 work has demonstrated the capability of retrieving information about the vertical distribution of
140 stratospheric aerosol from limb scatter measurements [*Bourassa et al.*, 2007, 2008a; *Rault and*
141 *Loughman*, 2007; *Tukiainen et al.*, 2008].

142 Our second means of comparison is with in situ measurements of aerosol size and
143 concentration from balloon-borne instruments that are launched three or four times a year from
144 Laramie, Wyoming (41.3°N, 105.7°W). Past use of this very long term data set in analyzing
145 volcanic aerosol layers in the stratosphere is well established [e.g., *Deshler et al.*, 2006]. We
146 suspect one of the main sources of discrepancy in *Kravitz et al.* [2010] was inaccurate estimation
147 of aerosol size, which would have a significant impact on our determination of aerosol optical
148 depth, as we describe in Section 3. Direct in situ measurements of aerosol particle size help us
149 address this hypothesis and provide additional useful data. We discuss these measurements in
150 more detail in Section 4.

151 Finally, compare the model results to data from multiple ground-based lidar stations. We
152 use measurements of aerosol optical depth and particle size, where available, from an elastic
153 backscattering lidar in Hefei, China (31.9°N, 117.1°E), two multi-wavelength aerosol Raman
154 lidars in Leipzig, Germany (51.4°N, 12.4°E) and Ny-Ålesund, Svalbard (78.9°N, 11.9°E), a lidar
155 in Halifax, Nova Scotia (44.6°N, 63.6°W) at Dalhousie University, and a lidar at the Mauna Loa
156 Observatory (19.5°N, 155.6°W). More description of these various instruments can be found in
157 Section 5. The locations of all of these data sources are shown in Figure 2.

158 The primary purpose of this paper is to explore the differences between modeled sulfate
159 aerosol optical depth and observed optical depth from the Sarychev eruption to analyze possible
160 sources of discrepancy between the two. A secondary purpose is to document the Sarychev
161 eruption with an extensive set of observations. We also want to continue the process of
162 comparison of the model results to the OSIRIS retrievals that was begun in *Kravitz et al.* [2010],
163 further showing indispensability of the OSIRIS measurements as a global atmospheric data
164 source.

165

166 **2. Climate Model**

167 To complete the climate modeling aspect of this study, we simulated the climate response
168 with a coupled atmosphere-ocean general circulation model. We used ModelE, which was
169 developed by the National Aeronautics and Space Administration Goddard Institute for Space
170 Studies [*Schmidt et al.*, 2006]. We used the stratospheric version with 4° latitude by 5° longitude
171 horizontal resolution and 23 vertical levels up to 80 km. It is fully coupled to a 4° latitude by 5°
172 longitude dynamic ocean with 13 vertical levels [*Russell et al.*, 1995]. The aerosol module
173 [*Koch et al.*, 2006] accounts for SO₂ conversion to sulfate aerosols, and the radiative forcing

174 (also called “adjusted forcing” in *Hansen et al.* [2005], which is the standard definition of
175 radiative forcing as adopted by the *IPCC* [2001]) of the aerosols is fully interactive with the
176 circulation. The dry aerosol radius is specified to be 0.25 μm , and the model hydrates these to
177 form a distribution with a median radius of approximately 0.30-0.35 μm , where aerosol growth is
178 prescribed by formulas in *Tang* [1996]. This distribution is consistent with the findings of
179 *Stothers* [1997], and was also used in the simulations of the eruptions of Katmai [*Oman et al.*,
180 2005] and Kasatochi [*Kravitz et al.*, 2010]. For more details on the specifications used in these
181 simulations, see *Kravitz et al.* [2010], which used the same modeling conditions.

182 Our control ensemble consisted of a 20-member collection of 4-year runs (2007-2010),
183 which involved increasing greenhouse gas concentrations in accordance with the
184 Intergovernmental Panel on Climate Change’s A1B scenario [*IPCC*, 2007]. No temperature
185 trend resulting from model spin-up was detected, due to corrective efforts utilizing previously
186 run initial conditions and sufficient tuning.

187 To examine the effects of the volcanic eruptions, we used a 20-member ensemble of 4-
188 year simulations covering the same time period. In these runs, greenhouse gas concentrations
189 increased in the same manner as in the control runs. We also injected 1.5 Tg of SO_2 into the grid
190 box centered at 52°N, 172.5°W, distributed equally in the three model layers that cover an
191 altitude of 10-16 km, on 12 June 2008. We recognize that the coordinates, amount, and year
192 used in this modeling study are not the same as the actual eruption. The reason for choosing
193 these particular values is to compare our simulations with those of the eruption of Kasatochi
194 Volcano on 8 August 2008 for which these specifications are valid [*Kravitz et al.*, 2010; *Kravitz*
195 *and Robock*, 2010]. Due to the distribution of the sulfate aerosols by the general circulation of
196 the atmosphere, our choice of spatial coordinates in simulating the eruption will not affect the

197 results. Also, the difference in atmospheric composition in the model between the years 2008
198 and 2009 is negligible, and any differences in results would be due to noise. We have adjusted
199 the labeling in our figures to make the eruption appear as if we simulated it in 2009, and for the
200 reasons we discuss here, this will not be detrimental to our conclusions. According to *Haywood*
201 *et al.* [2010], the results of which appeared after we completed our model runs, the simulations
202 reflect an incorrect choice of the amount of SO₂ that was injected into the lower stratosphere.
203 We address this later when we discuss the discrepancy between our modeled results and the
204 observations of aerosol optical depth.

205 ModelE has been shown to be realistic in simulating past volcanic eruptions. Simulations
206 of the climate response to volcanic eruptions with this model have been conducted for the
207 eruptions of Laki in 1783-1784 [*Oman et al.*, 2006a, 2006b], Katmai in 1912 [*Oman et al.*,
208 2005], and Pinatubo in 1991 [*Robock et al.*, 2007]. In all of these cases, ModelE simulations
209 agreed with observations and proxy records to such a degree that we are confident in this
210 model's ability to predict the climatic impact of volcanic eruptions, meaning model
211 representation of aerosol optical depth is accurate. *Kravitz et al.* [2010] also found the temporal
212 and spatial patterns of optical depth generated by ModelE to be consistent with those measured
213 by OSIRIS.

214

215 **3. Aerosol Optical Depth: Model vs. OSIRIS**

216 *Kravitz et al.* [2010] performed an extensive comparison between the modeled sulfate
217 aerosol optical depth and the retrievals obtained by OSIRIS. They encountered a discrepancy of
218 an order of magnitude, some of which was attributed to various assumptions made in both the

219 model and the radiative transfer of the satellite instrument. The eruption of Sarychev gives us
220 another opportunity to further investigate this discrepancy.

221 Figures 3 and 4 show the model calculations of the anomaly in spatial and temporal
222 extent of total sulfate aerosol optical depth (mid-visible, $\lambda = 550$ nm). Anomaly is defined as the
223 difference between the volcano ensemble and the control ensemble, thus removing the
224 contribution to optical thickness from tropospheric sulfate aerosols. Therefore, we refer to these
225 plots as volcanic sulfate aerosol optical depth. The largest anomaly of nearly 0.1 in Figure 3
226 occurs in August after the eruption. *McKeen et al.* [1984] report the chemical lifetime of SO_2 to
227 be 30-40 days, giving an *e*-folding lifetime of 10-14 days. However, the *e*-folding conversion
228 times for aerosols from the 1982 eruption of El Chichón and the 1991 eruption of Mt. Pinatubo
229 were 30-40 days [*Heath et al.*, 1983; *Bluth et al.*, 1992, 1997; *Read et al.*, 1983], giving a
230 chemical lifetime of 90-120 days. *Carslaw and Kärcher* [2006] also calculate an *e*-folding time
231 of the chemical conversion rate to be 30 days. The actual conversion rate depends on details
232 specific to each eruption, but this peak anomaly in August is consistent with these reported
233 values of chemical lifetime.

234 The bulk of the aerosol cloud does not pass south of 30°N , which is consistent with
235 *Stothers* [1996], although smaller values of sulfate optical depth are detectable in the Northern
236 Hemisphere tropics. Large scale deposition has removed most of the volcanic aerosols by
237 February after the eruption, with nearly all remnants disappearing before April. Radiative
238 forcing due to the sulfate aerosols becomes smaller in magnitude than -0.25 W m^{-2} well before
239 this time, dropping below this threshold even before winter.

240 Vertical profiles of stratospheric aerosol extinction were retrieved from the OSIRIS
241 measurements at a wavelength of 750 nm using the SASKTRAN forward model [*Bourassa et*

242 *al.*, 2008b]. Figure 5 shows a comparison between OSIRIS retrievals and climate model results,
243 divided into three latitude bins. In all latitude bins, background levels are very similar between
244 the model average and OSIRIS, with differences in τ within ± 0.002 . The OSIRIS background
245 levels may be slightly higher in the Arctic bin (70°N to 80°N) due to assumptions made in model
246 levels of sulfate aerosols, or perhaps the model has a slightly higher deposition rate than is found
247 in the atmosphere, resulting in a lower equilibrium level of background aerosol. We discuss later
248 deposition rates from the eruption in more detail. The model average is higher in June in the
249 middle bin (50°N to 60°N) than the OSIRIS retrievals because the model output is given in
250 monthly averages, and by late June, some of the aerosols due to Sarychev would already have
251 formed.

252 In the middle bin, peak optical depth occurs in late July, approximately the same time in
253 both the model and OSIRIS retrievals. This implies an SO₂ chemical lifetime of approximately
254 40-50 days, which is in line with the results of *McKeen et al.* [1984]. Table 1 shows the
255 comparison of decay in optical depth. The model tends to have autumn deposition rates that are
256 higher than are measured by OSIRIS, based on a linear fit of the data. However, in the Arctic
257 bin, peak optical depth occurs much later for the model, and in the near-tropical bin (20°N to
258 30°N), the peak occurs earlier. This is unlikely due to an incorrect conversion time from SO₂ to
259 sulfate, as a similar problem would be noticeable in all three bins. A likely candidate is
260 improperly calculated stratospheric circulation in the model, which distributes the aerosols to the
261 tropics slightly too quickly and to high latitudes too slowly. However, we are unable to
262 accurately diagnose the cause of this problem at this time.

263 Similar to the comparison of modeled and retrieved aerosol optical depth for Kasatochi in
264 *Kravitz et al.* [2010], the peak optical depth calculated by ModelE is nearly one full order of

265 magnitude larger than the retrievals obtained from OSIRIS in the Arctic bin and approximately 5
 266 times larger in the middle bin. In *Kravitz et al.*, several possible sources of discrepancy were
 267 outlined. One prominent source is the difference in wavelength used to calculate optical depth.
 268 ModelE calculates optical depth in the mid-visible ($\lambda=550$ nm), and OSIRIS retrieves in the near
 269 infrared ($\lambda=750$ nm). Since the radiative effects of the stratospheric aerosols follow an
 270 Ångstrom relationship, we would expect this to affect our results.

271 In ModelE, we assumed an aerosol dry radius of $0.25 \mu\text{m}$, consistent with past data from
 272 volcanic eruptions as found by *Stothers* [1997]. We used this value for the current set of
 273 simulations, and it was also used in the simulations of Kasatochi [*Kravitz et al.*, 2010] and
 274 Katmai [*Oman et al.*, 2005]. Based on ambient relative humidity values, aerosols of this initial
 275 size will increase in radius by at most 20-40%, according to formulas by *Tang* [1996]. These
 276 formulas are explicitly used in ModelE and are thus suitable for our calculations. This results in
 277 a hydrated aerosol median radius of $0.30\text{-}0.35 \mu\text{m}$.

278 According to the ModelE code, an aerosol radius of this size would have an Ångstrom
 279 exponent of $0.75\text{-}1.05$, resulting in OSIRIS retrievals being as little as 78% of ModelE results,
 280 based solely on using a different wavelength. More succinctly, the radiation code in ModelE
 281 calculates

$$283 \quad \frac{\text{AOD at } 750 \text{ nm and } r_{\text{dry}} = 0.25 \mu\text{m}}{\text{AOD at } 550 \text{ nm and } r_{\text{dry}} = 0.25 \mu\text{m}} \approx 0.78$$

284
 285 *Schuster et al.* [2006] and *Eck et al.* [1999] have measured Ångstrom exponents of this value to
 286 be consistent with the particle sizes that we have assumed in our simulations.

287 This alone does not fully explain the discrepancy between ModelE results and OSIRIS
288 retrievals. One additional source of error could be in assumed particle size. To properly
289 calculate optical depth, the model requires an assumption of particle size. Moreover, the model
290 assumes a unimodal gamma distribution, whereas reality may not have such a clearly defined
291 distribution. *Haywood et al.* [2010] indeed found two aerosol modes in a lognormal distribution:
292 an Aitken mode with effective radius 0.0065 μm and an accumulation mode of effective radius
293 0.095 μm . ModelE cannot model aerosols with a dry radius below 0.01 μm , so our model results
294 are incapable of capturing this smaller mode, although due to the very small size of these
295 particles, contributions to optical depth from the Aitken mode are likely not significant. Even in
296 the accumulation mode, the results of *Haywood et al.* suggest a gross overestimation of particle
297 size in our modeling study.

298 *Russell et al.* [1996] calculated a fit to a variety of measurements of aerosol effective
299 radius for the eruption of Pinatubo. In the first four months of the eruption, the effective radius
300 increased linearly from a background level of approximately 0.12 μm in May before the eruption
301 to 0.34 μm in September after the eruption, reaching a peak of 0.56 μm in April 1992. This is
302 not perfectly comparable with calculations of non-area-weighted radius, so a conversion must be
303 made. For a lognormal distribution, which is applicable to volcanic aerosols,

304

$$305 \quad r_{\text{eff}} = r_g \exp\left[\frac{5}{2}(\ln \sigma_g)^2\right]$$

306

307 where r_g is essentially the median radius, and σ_g is the distribution width. Although the eruption
308 of Pinatubo showed a clearly bimodal aerosol distribution structure for most of the aerosol
309 lifetime [*Russell et al.*, 1996], ModelE is only capable of representing a unimodal distribution, so

310 this is a good approximation. Because $(\ln \sigma_g)^2 \geq 0$, $\exp\left[\frac{5}{2}(\ln \sigma_g)^2\right] \geq 1$, meaning r_{eff} is always at
 311 least as large as r_g .

312 Simulations of the eruption of Pinatubo performed with ModelE [Oman *et al.*, 2006] used
 313 a dry radius of 0.35 μm , which results in a hydrated aerosol median radius of 0.47-0.52 μm .
 314 These results are consistent with *Stothers* [2001], but they are much higher than the observations,
 315 especially in the few months just after the eruption. This raises the possibility that, despite being
 316 consistent with pyrheliometric data, the model tends to overestimate aerosol size.

317 To capture this possibility, we performed the same calculations using ModelE's radiation
 318 code, but specifying a dry radius of 0.08 μm , which is approximately 1/3 our initial estimate of
 319 dry radius. This results in a hydrated aerosol radius of approximately 0.09-0.11 μm , again based
 320 on the formulas of *Tang* [1996]. We chose this radius to analyze the balloon-borne
 321 measurements of aerosol median radius, which are discussed in Section 4. This much smaller
 322 radius results in an Ångström exponent of approximately 2, as well as the relation

323

$$324 \frac{\text{AOD at 750 nm and } r_{\text{dry}} = 0.08 \mu\text{m}}{\text{AOD at 550 nm and } r_{\text{dry}} = 0.08 \mu\text{m}} \approx 0.38$$

325

326 This clearly shows the importance of an accurate estimate of the aerosol radius. An incorrect
 327 estimation of the aerosol radius in the model would mean a larger abundance of smaller particles
 328 and many fewer larger particles than the distribution we originally calculated. Using the Mie
 329 theory formulation of optical depth,

330

331
$$\tau = \int_0^{\infty} \int_0^{\infty} Q_{ext}(m, r) \cdot \pi r^2 \cdot N(r) dr dz$$

332
 333 this amounts to larger values of N for smaller values of r , and vice versa. Also, since scattering
 334 is more efficient for smaller particles, as r decreases, Q_{ext} increases. Determining the cumulative
 335 effect of these changes without re-running the model simulations is difficult due to the
 336 dependence of the shape of the aerosol distribution on the initial dry radius, as well as available
 337 humidity that can contribute to aerosol growth, which has a large dependence upon model
 338 dynamics. However, the effects of aerosol size alone can contribute another factor of 2-2.5
 339 beyond the estimates in *Kravitz et al.* [2010] of the discrepancy found in the Kasatochi
 340 comparison.

341 To some degree, particle size can also have a systematic impact on the OSIRIS results. In
 342 order to retrieve the aerosol extinction profile from limb scatter measurements the shape of the
 343 scattering phase function must be known or assumed. For the OSIRIS retrievals, a Mie code is
 344 used to calculate the scattering phase function for a log-normal particle size distribution. In this
 345 case, the OSIRIS retrievals are performed using the scattering phase function for a median, or
 346 mode, radius of 0.08 μm and a mode width of 1.6. Using the above definition, these values
 347 correspond to an effective radius of 0.14 μm . These are the same assumptions used for the
 348 OSIRIS retrievals of aerosol extinction following the Kasatochi eruption shown in *Kravitz et al.*
 349 [2010] and *Bourassa et al.* [2010]. As discussed in detail by Bourassa et al. [2007], uncertainty
 350 in the particle size distribution systematically affects the retrieved extinction. McLinden et al.
 351 [1999] show that for larger particle sizes, most likely in volcanically modified conditions, the
 352 phase function remains relatively stable at 750 nm, and systematic error remains on the order a
 353 few percent. However, for dramatically larger particle sizes the impact on the OSIRIS retrievals

354 could be as large as 30 or 40 percent adding an additional factor of uncertainty due to particle
355 size in the comparison between OSIRIS and the modeled optical depths.

356 Another reason explored in Kravitz et al. [2010] is the lower altitude level used to
357 calculated the stratospheric aerosol optical depths from the OSIRIS retrieved extinction profiles.
358 The lower bound is chosen to be the $\theta = 380$ K level of potential temperature. This assumption
359 is made to avoid attempting to retrieve extinction from clouds, dust, and other scattered signal
360 that are not stratospheric sulfate. However, using this as the lower bound for measurements has
361 the potential to reduce optical depth measurements, as OSIRIS will not account for aerosols
362 between the $\theta = 380$ K line and the true thermal tropopause. Figure 6 again shows optical depth,
363 taking into account this new lower bound, as well as combining the effects of the Ångström
364 exponent described above. Compared with Figure 4, optical depth in the midlatitudes and
365 subtropics is largely unchanged, with some areas of slight increase, indicating the thermal
366 tropopause is actually higher than the $\theta = 380$ K line. However, high latitude optical depth
367 patterns are much lower, sometimes by more than a factor of 2, indicating OSIRIS possibly
368 underestimates high latitude optical depth by assuming too high a base altitude for measurement.
369 Combining these results with scaling due to wavelength, as well as the possibility of using an
370 incorrect aerosol radius, gives the bottom left and top right panels of Figure 6.

371 *Haywood et al.* [2010] reported the upper tropospheric/lower stratospheric loading due to
372 Sarychev to be 1.2 Tg of SO₂. Although we were unable to obtain other firm estimations for this
373 value, this indicates our modeled aerosol optical depth values are overestimated by 25%. Arlin
374 Krueger [personal communication, 2010] estimated the atmospheric loading to be 1.5 Tg, which
375 was exactly his estimate of the loading due to Kasatochi. Kai Yang's group at NASA Goddard
376 Earth Sciences and Technology Center reported the atmospheric loading to be near 2.0 Tg SO₂

377 [A. Krueger, personal communication], which was the same value they reported for the eruptions
378 of Okmok and Kasatochi [Yang *et al.*, 2010]. Since the model results show higher optical depths
379 than the OSIRIS retrievals, we suspect the model overestimated the atmospheric loading, so, for
380 the purposes of calculating discrepancy, we will scale our model results by 0.8. The results of
381 this are shown in the bottom right panel of Figure 6. This panel shows that the maximum
382 overestimation of aerosol optical depth by the model due to these reasons is quite large, although
383 not as large as the overestimation of optical depth due to Kasatochi in Kravitz *et al.* [2010].

384 Figure 7 shows the combination of these three sources of error in comparison with
385 OSIRIS retrievals. When these potential errors are taken into account, the fit of the model to the
386 observations of the volcanic aerosols is quite good. Under this scaling, the fit to the background
387 level of stratospheric aerosols is very poor, which is expected, since the assumptions we made
388 regarding overestimation are specific to volcanic aerosols. Also, the mismatch of aerosol decay
389 rates becomes visibly clear. The decay rate in the summer appears to be good, although the
390 small amount of data is not conducive to the construction of a linear fit. However, as in Figure 5,
391 the autumn decay rate in the model appears to be larger than is observed. Also more apparent is
392 the peak in subtropical optical depth, which is much larger and much earlier than is observed.

393 Kravitz *et al.* [2010] discovered evidence for additional sources of discrepancy in their
394 comparison, some of which may also be relevant to the eruption of Sarychev. Although we
395 cannot quantify the degree to which they might affect our results, we can briefly discuss them.

396 One of the largest potential sources of discrepancy is that not all of the SO₂ may have
397 been injected above the tropopause, meaning some of the aerosols would have formed in the
398 troposphere and deposited very rapidly. This leaves the option that the model's overestimation
399 of SO₂ loading is even greater than is discussed above.

400 Additionally, as was found by *Schmale et al.* [2010] for the eruption of Kasatochi, not all
 401 of the volcanic aerosol layer is necessarily composed of sulfate, which will affect the radiative
 402 properties of the aerosol layer. *Schmale et al.* also discovered some SO₂ remained as late as
 403 three months after the eruption, possibly indicating overly rapid conversion of SO₂ into sulfate in
 404 the model. Both of these would indicate a potential source of additional overestimation of
 405 sulfate aerosol optical depth by the model.

406 Finally, some additional possible sources of discrepancy are related to possible inaccurate
 407 representations of removal processes in the model. The model can potentially have an incorrect
 408 rate of aerosol deposition, although our comparison in Section 3 suggests this is a negligible
 409 explanation of discrepancy. More unknown is the phase of the QBO and its effects on the
 410 removal efficiency, and the phase and magnitude of tropical modes, which we would not
 411 necessarily expect the model to accurately represent, given the large natural variabilities of these
 412 processes.

413

414 **4. Comparison using in situ aerosol profiles**

415 Our second means of comparison with model output is in situ aerosol measurements from
 416 balloon-borne instruments lofted from Laramie, Wyoming (41.3°N, 105.7°W). Use of this very
 417 long-term data source has been well established for both volcanic eruptions and background
 418 stratospheric aerosol concentrations [e.g., *Deshler et al.*, 2006]. The size resolved number
 419 concentration measurements are fit to either unimodal or bimodal lognormal size distributions of
 420 the form [e.g., *Hoffmann and Deshler*, 1991; *Deshler et al.*, 1993]

421

$$422 \quad n(r) = \sum_{i=1}^2 \frac{N_i}{\ln(\sigma_i)\sqrt{2\pi}} \cdot \frac{1}{r} \exp\left[-\frac{\ln^2(r/r_i)}{2 \cdot \ln^2(\sigma_i)}\right]$$

423

424 where N_i is aerosol number density, r_i is the aerosol median radius, and σ_i is the standard
425 deviation of the distribution. *Deshler et al.* [2003] provide more details on the specifics of the
426 measurements, their uncertainties, and the derivation of size distributions and their moments.
427 Measurement uncertainties lead to an error of the fits by $\pm 30\%$ for the median radius, $\pm 20\%$ for
428 the standard deviation, and $\pm 40\%$ for surface area and volume. In the aerosol measurements
429 following the Sarychev eruption, the larger aerosol mode has such a low number concentration
430 that the fit is effectively unimodal. *Deshler et al.* [1997] showed the Pinatubo aerosols
431 developed a clearly bimodal structure approximately 40 days after the eruption, so perhaps the
432 Sarychev eruption did not eject enough material to create this larger mode.

433 Figure 8 shows in situ measurements from 22 June 2009, ten days after the initial
434 eruption of Sarychev. For comparison, it also shows results from 3 July 2007, over a year after
435 Soufriere Hills and prior to Kasatochi. This sounding was chosen because it was approximately
436 the same time of year as the 22 June 2009 sounding, has a similar temperature profile, and was a
437 relatively clean period for volcanic eruptions. We chose a sounding within close temporal
438 proximity to 2009, as the stratospheric aerosol layer has become increasingly thick since
439 approximately 2000, so only recent soundings would be suitable for comparison [*Hofmann et al.*,
440 2009].

441 The 2009 measurements show no significant differences from the 2007 measurements. If
442 the chemical lifetime of SO_2 for this eruption is on the lower end of the estimates given in the
443 previous section, similar to the values reported by *McKeen et al.* [1984], then a significant
444 amount of aerosols from Sarychev would have been formed by 22 June 2009. Moreover, back-

445 trajectory calculations show the volcanic plume could have reached Laramie by this time
446 [*Haywood et al.*, 2010].

447 Radiosondes are launched every 12 hours from Sakhalin Island (47.0°N, 142.7°E), which
448 is very close to the eruption site of Sarychev (48.1°N, 153.2°E). The initial plume height of 11-
449 16 km [*Haywood et al.*, 2010] corresponds to a potential temperature range of 342-400 K,
450 according to radiosonde data from 00Z 16 June 2009 [*Durre et al.*, 2006]. This station is
451 southwest of the eruption site, so this result should not have been altered by the eruption, due to
452 the predominating westerlies at this latitude. Due to the stratosphere's inherent stability,
453 stratospheric motion is often confined to isentropic layers [*Holton*, 2004]. Although cross-
454 isentropic motion is possible, due to diabatic heating or lofting of the isentropes due to the
455 pressure wave of the volcanic eruption, it is plausible that the volcanic plume remained confined
456 to this range of potential temperatures through its passage over Laramie. The 22 June 2009
457 sounding reports the potential temperature range of 342-400 K corresponds to an altitude range
458 from below the tropopause up to 16 km. Therefore, it is unlikely that the measurements from 22
459 June 2009 show any aerosols from the Sarychev eruption, as these altitudes show little difference
460 from background levels.

461 Figure 9, similar to Figure 8, shows results from measurements on 7 November 2009,
462 five months after the eruption, and from 17 October 2005. The 2005 measurements were chosen
463 because 2005 was a quiescent year for stratospheric aerosols, yet according to *Hofmann et al.*
464 [2009], was still close enough in time to the eruption to have comparable levels of background
465 stratospheric aerosol, and the time of year and tropopause heights were similar in both profiles.
466 The aerosols have had time to age since the June sounding, resulting in much larger volumes and
467 surface areas. The aerosols have also settled, which is evidenced by a large area of increased

468 volume and surface area from the tropopause (13.0 km at this time and latitude) to 19.0 km in
469 altitude, with a strong peak at 14.0 km. The reported median radius at 14 km in altitude is
470 approximately 0.07-0.08 μm , which motivates our choice of radius in the calculations in Section
471 3.

472 *Deshler et al.* [1997] calculated a subsidence rate in the Southern Hemisphere of 3-4 km
473 a^{-1} for the stratospheric aerosols from Pinatubo, which is consistent with the fall rate of a particle
474 of radius 0.5 μm . This is likely much larger than the Sarychev aerosols, implying that
475 gravitational settling mechanisms would result in a much slower fall speed for the Sarychev
476 aerosols. However, assuming the Pinatubo deposition rate for the eruption of Sarychev, the
477 aerosol plume would have descended no more than 1.5-2 km over the period June to November
478 and 2.5-3 km over the period June to March. Therefore, to explain the large peaks in Figure 9 at
479 14.0 km, the initial plume height cannot have been greater than 16.0 km in altitude. This is again
480 consistent with the results in *Haywood et al.* [2010]. However, if the initial plume height were
481 16.0 km, at the same deposition rate, the aerosols could not have descended below 13.0 km by
482 March 2010. The tropopause height in March was measured to be 11.0 km, and no significant
483 stratospheric aerosol layers were detected at this time in the model results or any of the data
484 sources, meaning all aerosols had been deposited out of the atmosphere and thus must have
485 descended lower than this height. Thus, assuming a rate of deposition identical to the Pinatubo
486 rate is contrary to our findings, meaning it is likely that the Sarychev aerosols have a higher
487 deposition rate than the Pinatubo aerosols.

488 This faster deposition rate can be explained by a number of factors. A large part of the
489 atmospheric lifetime of stratospheric aerosols is poleward transport, where large scale descent of
490 air in the winter is responsible for removal of the aerosols [e.g., *Hamill et al.*, 1997]. If the

491 aerosols already begin at high latitudes, as in the case of Sarychev, the absence of the need for
492 poleward transport will necessarily decrease the atmospheric lifetime. *Oman et al.* [2005]
493 obtained similar results in their simulations, as they found an *e*-folding lifetime of 1 year for
494 aerosols from Pinatubo, a tropical eruption, and 8-9 months for aerosols from Katmai, a high
495 latitude eruption. Moreover, a large part of the aerosol plume from Sarychev is concentrated in
496 the midlatitude storm tracks, where tropopause folding is responsible for even more removal of
497 stratospheric aerosols [e.g. *Kravitz et al.*, 2009]. Finally, the relatively small amount of aerosols
498 created was insufficient to avoid these deposition factors, meaning very little aerosol remained in
499 the stratosphere by the following spring. Conversely, Pinatubo was a very large eruption,
500 injecting gases and particles to much higher altitudes, and thus aerosols remained in the
501 stratosphere for multiple years afterwards. The processes controlling aerosol deposition at higher
502 altitudes may be weighted significantly different than processes near the tropopause where
503 dynamics is more of a factor. These differences may account for the calculation of a slower
504 deposition rate from Pinatubo [*Deshler et al.*, 1997].

505 Mie theory was used to calculate aerosol extinction profiles and optical depth at 758 nm
506 from the in situ aerosol profiles on 17 October 2005, 22 June 2009, and 7 November 2009,
507 Figure 10. The profiles and optical depths on 17 October and 22 June are quite similar. In
508 contrast, the 7 November 2009 sounding shows a stratospheric optical depth of 0.0044, over
509 three times higher than observed earlier. The increase in optical depth on 7 November 2009 is
510 from an increase in aerosol between the tropopause and 20 km. This increase in aerosol optical
511 depth by more than a factor of three is due to the eruption of Sarychev. OSIRIS measurements
512 for the latitude bin 40°N-45°N and for the week of 7 November 2009 give an optical depth of
513 0.0109, which is over a factor of two greater than the in situ measurements. This is nearly within

514 the in situ measurement error of $\pm 40\%$ which applies to any aerosol moment calculated. After
515 accounting for the sources of discrepancy we discuss in Section 3, as well as the uncertainty in
516 the in situ measurements, the model and in situ measurements are relatively similar. The
517 uncertainty is too large for us to reliably determine the degree to which they disagree.

518

519 **5. Further comparison using lidar data**

520 To better characterize our results, our simulations will be compared with observations
521 from five ground-based lidar sources in Hefei, China (31.9°N, 117.1°E), Leipzig, Germany
522 (51.4°N, 12.4°E), Ny-Ålesund, Svalbard (78.9°N, 11.9°E), Halifax, Nova Scotia, Canada
523 (44.6°N, 63.6°W), and Mauna Loa, Hawaii (19.5°N, 155.6°W) (Figure 2).

524 The lidar in Hefei is an elastic backscattering lidar for profiling aerosol backscatter
525 coefficient at 532 nm based on a Nd:YAG laser with a second harmonic generator. Aerosol
526 coefficient profiles below about 25 km above ground level were derived from lidar data using the
527 Fernald method with an assumed lidar ratio of 50 sr.

528 The results from this lidar (Figure 11) show a peak in backscatter in September 2009 at
529 an altitude of 18-19 km, which corresponds to an aerosol optical depth of approximately 0.014.
530 All profiles are very similar above 21 km in altitude, suggesting this as an upper bound for the
531 plume height. July 2009 shows a slight peak, whereas the profile for June 2009 is nearly
532 identical to months prior. All backscatter profiles from December 2009 onward are similar to the
533 background. However, aerosol optical depth measurements from 2010 are slightly larger than in
534 early 2009, prior to the eruption, suggesting a small amount of aerosol remained in the
535 stratosphere through at least the winter following the eruption.

536 Although we would not expect the model to perfectly capture the distribution of the
537 aerosol plume, the aerosol optical depth measurements at Hefei are of similar magnitude to the
538 climate model results. The lidar measures a peak optical depth of 0.014 in September, whereas
539 the model calculates a peak of 0.012 in August. This is consistent with our comparison with
540 OSIRIS, in that the modeled peak optical depth at this latitude occurs earlier than is observed.
541 The autumn deposition rate also appears to be higher in the model than the observations. The
542 altitude of reported peak backscatter is at a similar altitude to peak aerosol retrievals as seen in
543 the 7 November 2009 in situ measurements. Optical depth measurements from the lidar are
544 slightly higher than in situ calculations, but the difference is within the range of uncertainty.

545 MARTHA (Multiwavelength Atmospheric Raman lidar for Temperature, Humidity, and
546 Aerosol profiling), a multiwavelength Raman lidar in Leipzig, Germany, has been in operation
547 since 1996 [Mattis *et al.*, 2010]. From it, we can obtain vertical profiles of the particle
548 backscatter coefficient at the three wavelengths of 355, 532, and 1064 nm, the extinction
549 coefficient at 355 and 532 nm, the corresponding lidar ratio at 355 and 532 nm, and profiles of
550 depolarization ratio at 532 nm. Mattis *et al.* [2002a, 2002b] and Ansmann *et al.* [2002] describe
551 in more detail the current system in operation, as well as error analysis. This lidar has been used
552 to evaluate the aerosol cloud resulting from past volcanic eruptions, including Pinatubo [Mattis,
553 1996; Ansmann *et al.*, 1997] and Kasatochi [Mattis *et al.*, 2010]. It has also had success in
554 retrieving aerosol microphysical properties [Wandinger *et al.*, 1995; Müller *et al.*, 1999].

555 The results from this lidar (Figure 12) show optical depth measurements about a factor of
556 2 lower than model results but approximately a factor of 2 higher than the in situ measurements.
557 The peak value of approximately 0.025 occurs in late July and mid August, which is 2-4 weeks
558 later than modeled peak optical depth. This factor of 2 can be explained by several potential

559 reasons. The spatial distribution of the volcanic plume in the model would not be expected to
560 perfectly match the lidar observations, especially considering the coarse spatial resolution of the
561 model. Also, several assumptions in both the model and observations could alter the results,
562 including the assumed lidar ratio of approximately 38 in determining optical depth, the base
563 altitude from which backscatter is integrated, an inaccurate estimation of the eruption size and
564 particle radius (as was discussed in Section 3), and in situ measurement uncertainty (as discussed
565 in Section 4). With the exception of those factors detailed in Sections 3 and 4, we are unable to
566 accurately quantify the degree to which our comparison is affected. Aerosol optical depth
567 returns to near background levels by December following the eruption.

568 To partly resolve discrepancies between this lidar and OSIRIS, Figure 13 shows the same
569 backscatter results as Figure 12, but optical depth is recalculated at 750 nm, using both the
570 thermal tropopause and the 380 K potential temperature line as the lower bound for integration.
571 Comparing with Figure 7, making these corrections still results in optical depth calculations that
572 are of the same order of magnitude as the OSIRIS retrievals and the corrected model output.
573 However, the differences between these corrections and the values in Figure 12 are rather small.

574 The Koldewey Aerosol Raman Lidar (KARL) is part of the AWIPEV research base in
575 Ny-Ålesund, Svalbard (78.9°N, 11.9°W, www.awipev.eu) and in operation since 2001. The light
576 source is a Nd:YAG laser, which transmits pulses at the three wavelengths of 355, 532, and 1064
577 nm at a repetition rate of 50 Hz. With a 70-cm telescope elastic backscattering at those three
578 wavelengths as well as N₂ and H₂O Raman signals and the depolarization ratio at the two shorter
579 wavelengths are detected. Backscatter coefficient profiles are calculated using the Klett method
580 with different lidar ratios [Klett, 1981]. KARL has mainly been used for characterizing the Arctic
581 spring troposphere, where Arctic haze occurs [Ritter *et al.*, 2004; Hoffmann *et al.*, 2009]. In

582 recent years, stratospheric volcanic aerosols, e.g. from the Kasatochi volcano [*Hoffmann et al.*,
583 2010] have also been observed.

584 The results from KARL (Figure 14) agree very well with the model simulations.
585 Modeled optical depth values and decay rates are nearly identical to the lidar retrievals.
586 Measured peak optical depth occurs in late July, which is earlier than the August peak in the
587 model. This is also consistent with the comparison with OSIRIS, in which modeled optical
588 depth peaked later than measured optical depth at this latitude. However, the maximum sulfate
589 aerosol optical depth of above 0.08 is found in August above Spitsbergen, which agrees with the
590 model results. The temporal variability of backscatter ratios and hence aerosol optical depth is
591 very high within the first 2 months after the volcanic eruption, due to the occurrence of several
592 distinct layers of enhanced backscatter (Figure 15). In September, stratospheric aerosol optical
593 depth was still high with 0.04 but less variable, due to a more uniform distribution of the sulfate
594 aerosols within the stratosphere. The temporal evolution of aerosol optical depth shown in
595 Figure 14 matches the model output for the Arctic bin in Figure 5. As stated earlier, these values
596 are much higher than the aerosol optical depths obtained with OSIRIS but could be confirmed by
597 co-located sun photometer measurements. These values are higher than the in situ measurements
598 by approximately one order of magnitude, but the comparability of these two sources of
599 measurement is uncertain, due to the large difference in latitude between the two sites.

600 The Dalhousie Raman Lidar is operated in Halifax, Nova Scotia, Canada (44.6°N, 63.6°W)
601 and measures vertical profiles of atmospheric scattering. The instrument employs a frequency-
602 doubled ND:YAG laser which transmits pulses of 532 nm wavelength light into the atmosphere
603 at a repetition rate of 20 Hz. The receiver consists of a 25-cm telescope and photomultipliers
604 with fast counting electronics to detect the signals. Profiles of the aerosol backscatter cross-

605 section are derived from the measured elastic lidar signals using the Klett Inversion technique
606 [Klett, 1981], assuming a constant lidar ratio of 40 sr for stratospheric aerosols. A more detailed
607 description of the instrument and aerosol optical property retrievals can be found in *Bitar et al.*
608 [2010].

609 The results for the lidar in Halifax (Figure 16) show peak backscatter in July of very
610 similar values to peak backscatter in the Leipzig lidar results. The altitudes of this peak
611 backscatter are more concentrated, ranging between 14-16 km for the Halifax results and 12-16
612 km for the Leipzig results. Also, the peak occurs approximately one month earlier than the
613 Leipzig measurements. These altitude ranges are consistent with model input, the findings of
614 *Haywood et al.* [2010], and the in situ measurements discussed in Section 4. Backscatter is near
615 background levels for the June and December measurements. Calculations of optical depth show
616 a peak of approximately 0.02, again in July, with a lower peak in August. This pattern matches
617 the model output quite well, although the modeled values of optical depth are approximately a
618 factor of 2 larger than the retrievals. The decay rate of optical depth also matches between the
619 two sources. The in situ measurements in November are approximately one order of magnitude
620 higher than the lidar measurements, but we are unable to determine what caused this large
621 discrepancy.

622 The NOAA Mauna Loa Observatory lidar uses a 30 Hz Nd:YAG laser producing the 1064
623 nm and 532 frequency-doubled wavelengths. The power at each wavelength is about 15 W, and
624 two 61-cm diameter mirrors are used to collect the scattered light. Photon-counting
625 photomultiplier tubes are used for both wavelengths and are electronically gated when needed.
626 The data acquisition electronics has 300 m altitude resolution, and files are normally saved every
627 5.6 minutes. The molecular signal is usually normalized in the interval from 35 to 40 km. The

628 molecular profile is derived from the Hilo radiosonde and a Mass Spectrometer Incoherent
629 Scatter model for the upper stratosphere. The error due to the signal statistics is about 5%. The
630 lidar is a primary instrument of the Network for the Detection of Atmospheric Composition
631 Change.

632 Figure 17 shows weekly observations from the Mauna Loa Observatory. The lidar
633 detected aerosols from the Sarychev eruption as early as July 1, at which time the aerosol cloud
634 remained confined to 14-16 km in altitude, which is similar to the in situ measurements.
635 Throughout its lifetime, the plume rose in altitude and spread to an altitude of approximately 16-
636 23 km, which is still the lower stratosphere in the tropics. The plume ceased to be detectable by
637 February, 2010. Modeled optical depth at this latitude shows a large peak in August of nearly
638 0.01, whereas the lidar shows a rather consistent optical depth, reaching a slight peak of 0.004.
639 Modeled optical depth also decays much more rapidly, showing very low levels by November,
640 2010, whereas the lidar detected aerosols for a few months after. The in situ measurements agree
641 with the lidar measurements quite well, showing a difference of approximately a factor of 2,
642 which is within the uncertainty range.

643 In general, the model results show agreement with the lidar retrievals, with differences in
644 aerosol optical depth being at most a factor of 2, with the exception of the Mauna Loa lidar,
645 where the differences were somewhat larger and the timeseries of optical depth values have very
646 different shapes. The Hefei lidar and the Svalbard lidar, both in agreement in magnitude with the
647 model results, were far in latitude from the original eruption site and assumed a lidar ratio of
648 approximately 50 sr. In contrast, the Leipzig and Halifax lidars were closer to the same latitude
649 as the eruption and assumed a lower lidar ratio of approximately 40 sr. Thus, two likely
650 explanations for the discrepancies in difference between lidar optical depth and model optical

651 depth are a difference in lidar ratio and a difference between modeled stratospheric circulation
652 and the real world. The factor of 2 can be explained by the same mechanisms as were discussed
653 in Section 3, i.e., an overestimation of particle size, the lidars' assumed higher base of integration
654 to avoid contamination of the measurements by cirrus clouds, and the amount of SO₂ injected in
655 the model. The larger disagreement with the Mauna Loa observatory is similar to our findings
656 for OSIRIS, in that for this particular eruption, the model did not accurately calculate optical
657 depth for the tropics. The in situ measurements agreed quite well with OSIRIS and
658 measurements from three of the lidar sites, showing at most a difference of a factor of 2, which
659 can be explained by measurement uncertainties and predicted sources of discrepancy.

660

661 **6. Discussion and Conclusions**

662 We evaluated ModelE simulations of the Sarychev volcanic plume using several different
663 observational data sets. In so doing, we discovered several areas in which ModelE could be
664 improved. The model has issues with stratospheric circulation, specifically the latitudinal spread
665 of the aerosols. We also found that the model deposits the aerosols out of the atmosphere too
666 quickly during autumn and winter.

667 We found that, however, after accounting for expected sources of discrepancy, the model
668 results and all reported sources of data show good agreement. Due to the difficulty of
669 determining the degree to which different wavelengths of measurement affect discrepancies
670 between OSIRIS and the lidar data, we are currently unable to make a thorough comparison of
671 the OSIRIS observations with the model results. However, intercomparison with the model
672 results and the lidar data suggests that OSIRIS is an accurate, useful means of obtaining
673 stratospheric aerosol optical depth.

674 Despite the agreement among our different sources of data, volcanic observation systems
675 still require a great deal of improvement. A range of reported amount of SO₂ injected into the
676 stratosphere, if used to force a climate model, would result in a large range of predicted climate
677 effects. Moreover, estimates of aerosol particle size are very sparse. As we discuss in Section 3,
678 accurate measurement of particle size, both initially and as the aerosols age, are essential to
679 accurate determining the radiative effects.

680
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891 **Table 1.** Results from the linear fit to optical depth data shown in Figure 5. The annual decay
 892 rate of optical depth in the model is approximately 5-7 times the decay rate measured by
 893 OSIRIS.

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 895 **(a) OSIRIS**
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Bin	Decay Rate (τ a⁻¹)	R²	Decay Rate calculated from Figure 5 (log₁₀(τ) a⁻¹)	R²
70°N – 80°N	0.0329	0.58	1.9003	0.59
50°N – 60°N	0.0178	0.48	1.0607	0.50
20°N – 30°N	0.0045	0.10	0.3833	0.10

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899 **(b) ModelE** (linear fit not plotted)

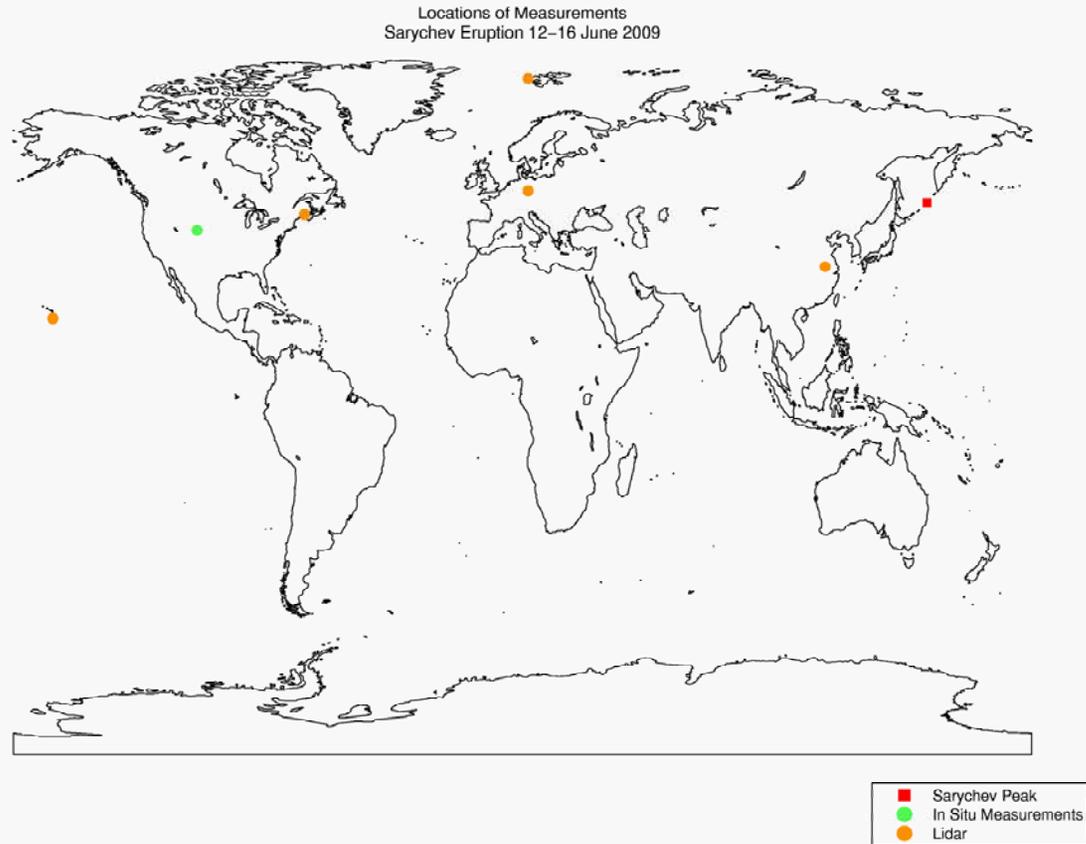
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Bin	Decay Rate (τ a⁻¹)	R²	Decay Rate calculated from Figure 5 (log₁₀(τ) a⁻¹)	R²
70°N – 80°N	0.1707	0.95	2.6730	> 0.99
50°N – 60°N	0.1303	0.97	2.7113	0.99
20°N – 30°N	0.0201	0.90	2.3871	0.81



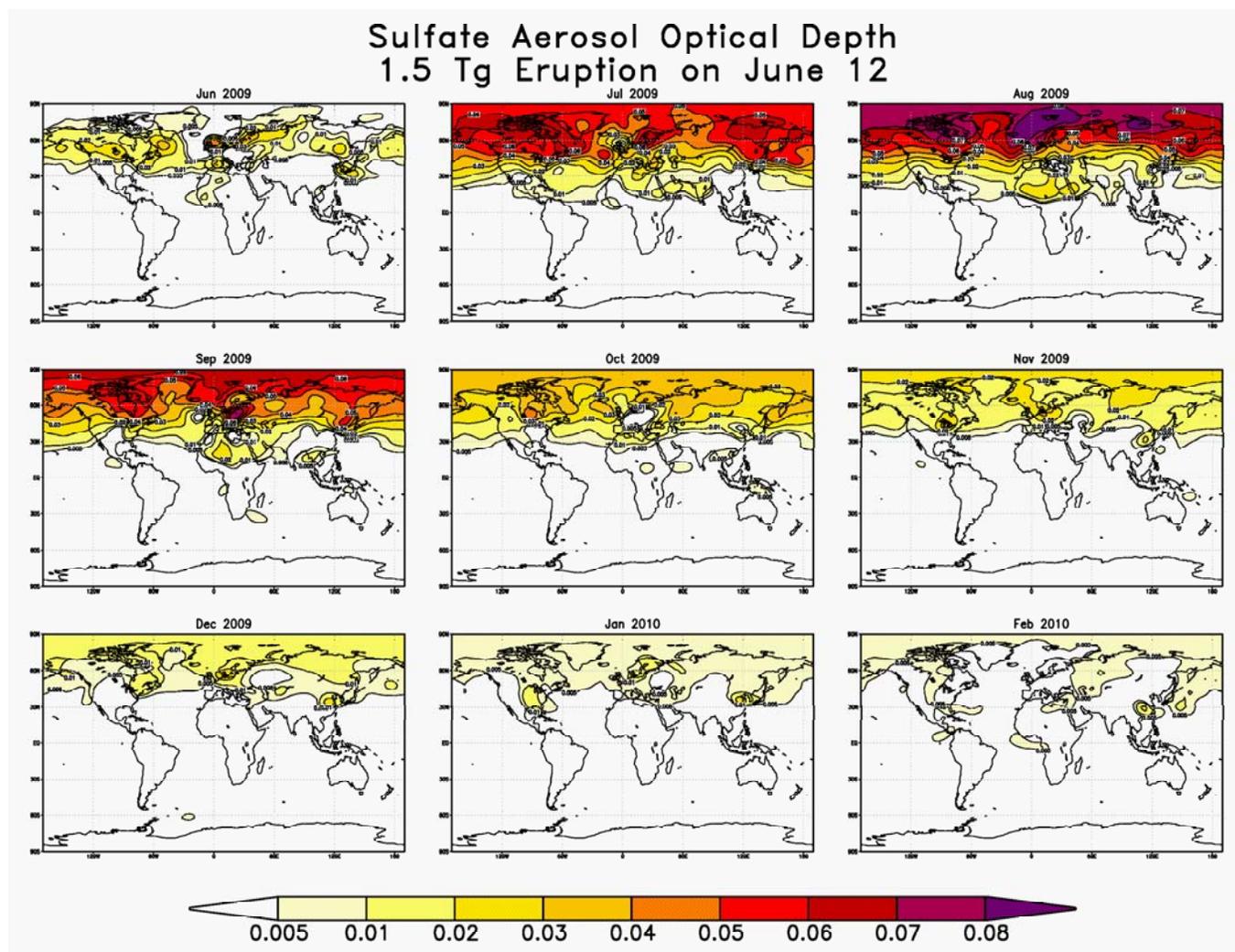
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902 **Figure 1.** The eruption of Sarychev volcano on 12 June 2009 as seen from the International Space Station [NASA, 2009]. Image
903 courtesy of the Image Science & Analysis Laboratory, NASA Johnson Space Center.



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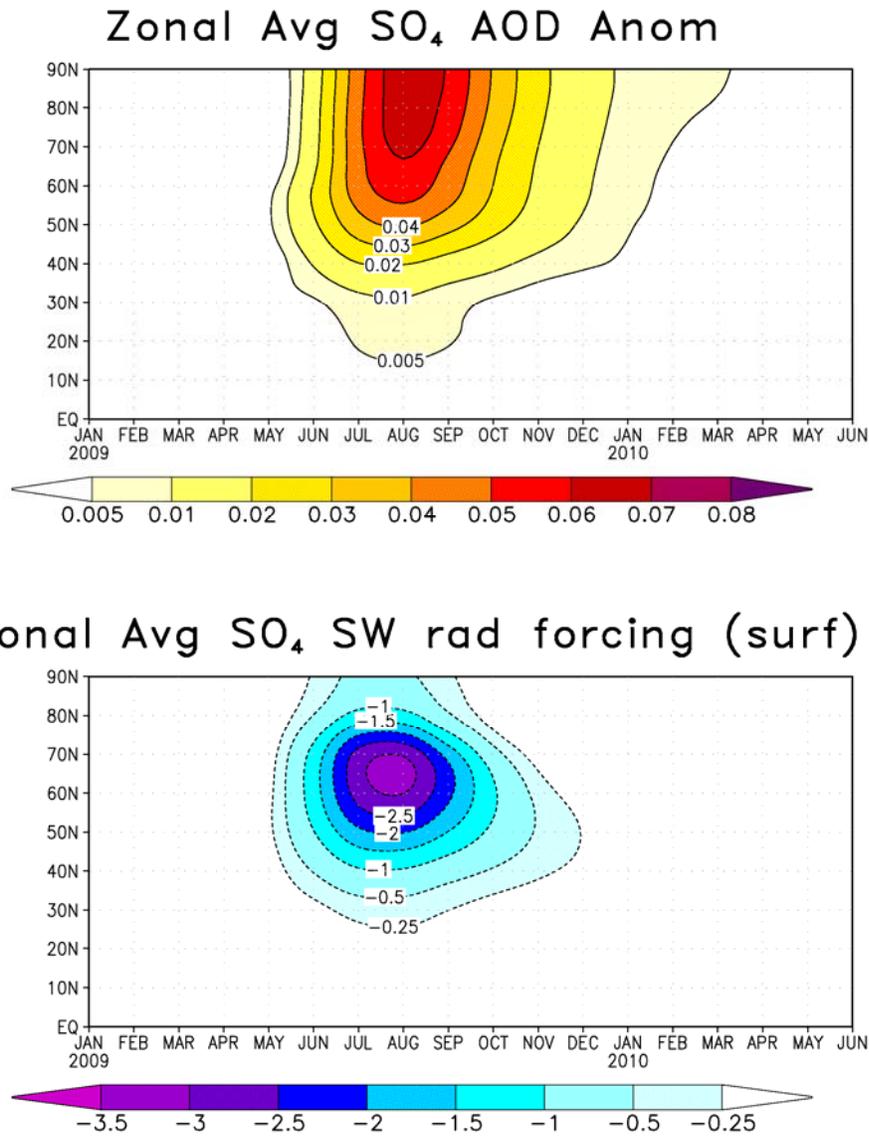
Figure 2. The locations of all point measurements used in our discussion of the Sarychev eruption. The site of the eruption is indicated by a red square. The in situ measurements from Laramie are indicated by a green dot. Lidar stations are indicated by orange dots. OSIRIS is a global measurement, so it cannot be included in this figure.



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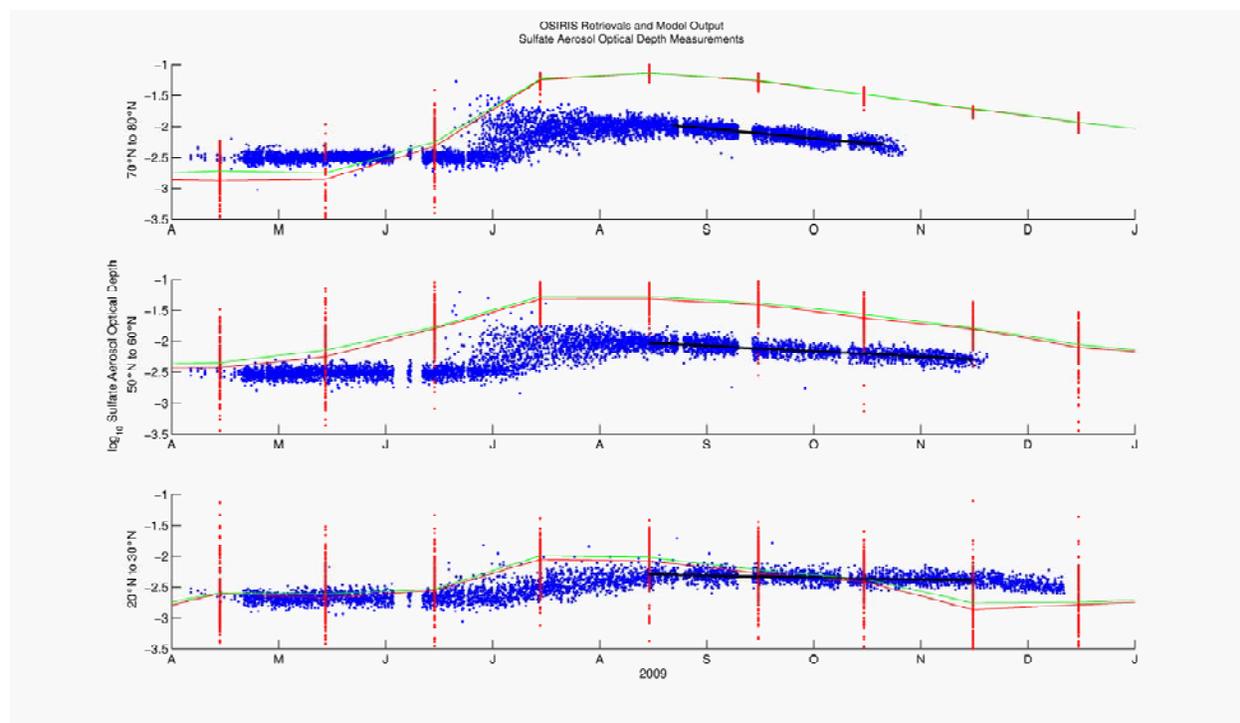
910 **Figure 3.** Time progression of anomaly in stratospheric sulfate aerosol mid-visible optical depth for the eruption of Sarychev from
 911 June 2009 to February 2010. Both the volcano ensemble and the baseline ensemble are averages of 20 runs. By February 2010,
 912 volcanic aerosols remaining in the atmosphere are at very low levels.

1.5 Tg Eruption on June 12



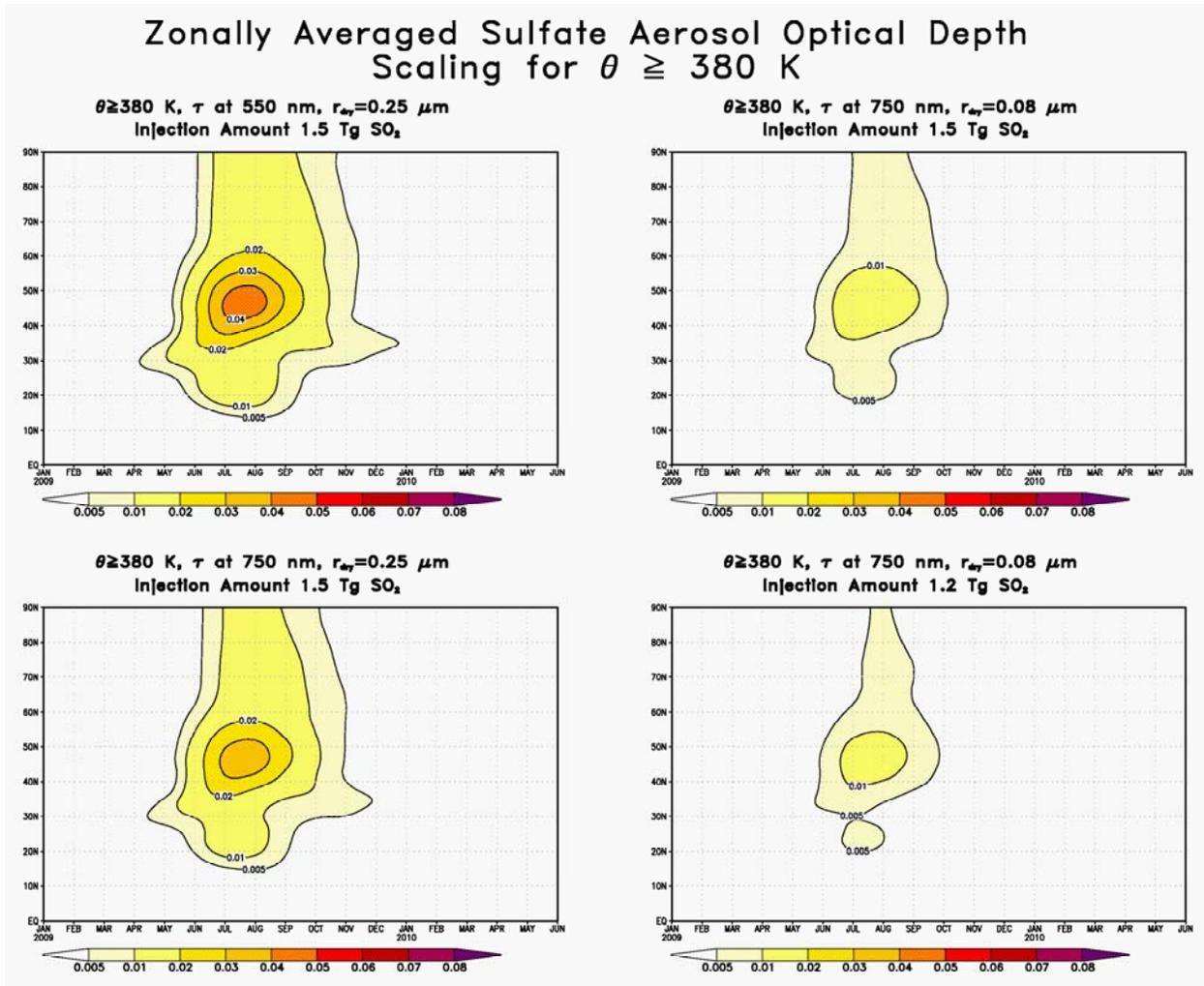
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914 **Figure 4.** Zonally averaged anomalies in stratospheric sulfate aerosol mid-visible optical depth
 915 and clear sky shortwave radiative forcing (W m^{-2}) at the surface due to sulfate aerosols. Only the
 916 Northern Hemisphere values are plotted, as the Southern Hemisphere values are zero. Results
 917 shown are for model simulations of the Sarychev volcanic eruption. Both the volcano ensemble
 918 and the baseline ensembles are averages of 20 runs. Results shown here are similar to those in
 919 Figure 3, i.e., most of the sulfate aerosols have been deposited out of the atmosphere by
 920 February, 2010. Radiative forcing due to the sulfate aerosols ceases to be detectable even
 921 sooner.



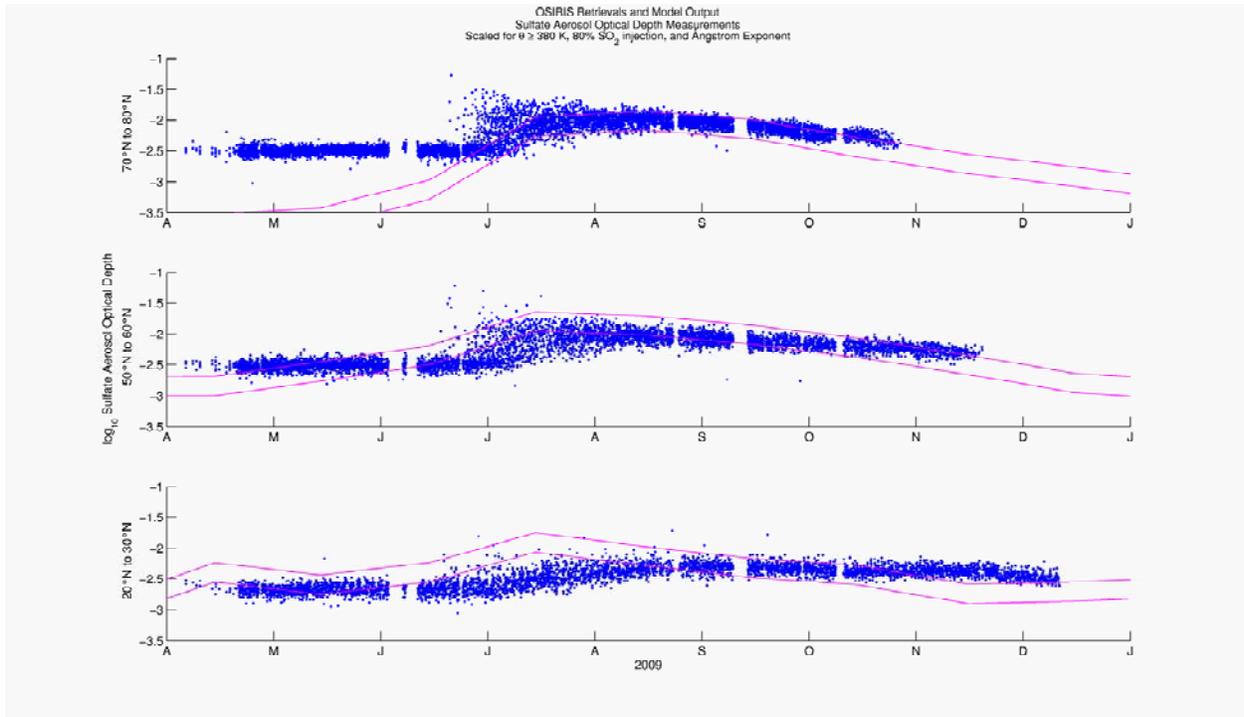
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Figure 5. Total stratospheric aerosol optical depth measured by OSIRIS at 750 nm and model results of optical depth at 550 nm. The month labels indicate the beginning of each month. All blue values are individual retrievals from OSIRIS, divided into three latitude bands. All red dots are individual grid box measurements of aerosol optical depth for each latitude band (72 for each latitude that falls into the above bands). The model output is placed on the 15th of each month, as these values represent monthly averages. The red line is an average of all red points (\log_{10} is taken after averaging), indicating an average of model optical depth in the given latitude band. The green line is the median of all red points. Black lines are linear fits to aid in understanding atmospheric deposition rates, the details of which are in Table 1. For OSIRIS measurements, the vertical column extends only from the 380 K level of potential temperature to 40 km altitude. OSIRIS coverage of the Arctic is not available from November to March due to the lack of sunlight.



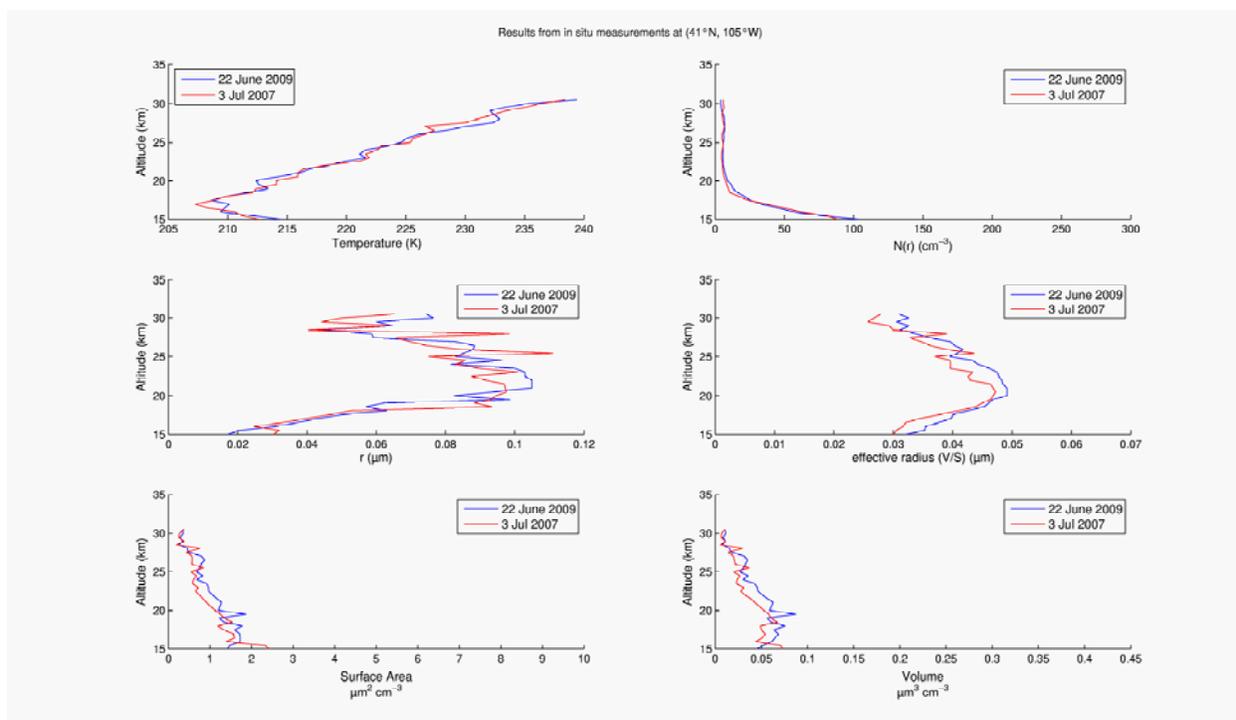
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Figure 6. Zonally averaged total stratospheric aerosol optical depth anomaly as calculated by the model. Top left shows anomaly in zonally averaged optical depth, scaled using the $\theta = 380$ K line as the tropopause instead of the thermal tropopause. Bottom left shows the same field multiplied by 0.78 to reflect the difference in measured optical depth due to a change in wavelength, assuming a dry radius of $0.25 \mu\text{m}$. Top right is again scaled using the $\theta = 380$ K line as the tropopause and is also multiplied by 0.38 to reflect the difference in measured optical depth due to a change in wavelength, assuming a dry radius of $0.08 \mu\text{m}$. Bottom right is the same as the top right but multiplied by 0.8 to reflect our overestimation of the initial SO_2 loading, which should have been 1.2 Tg instead of 1.5 Tg .



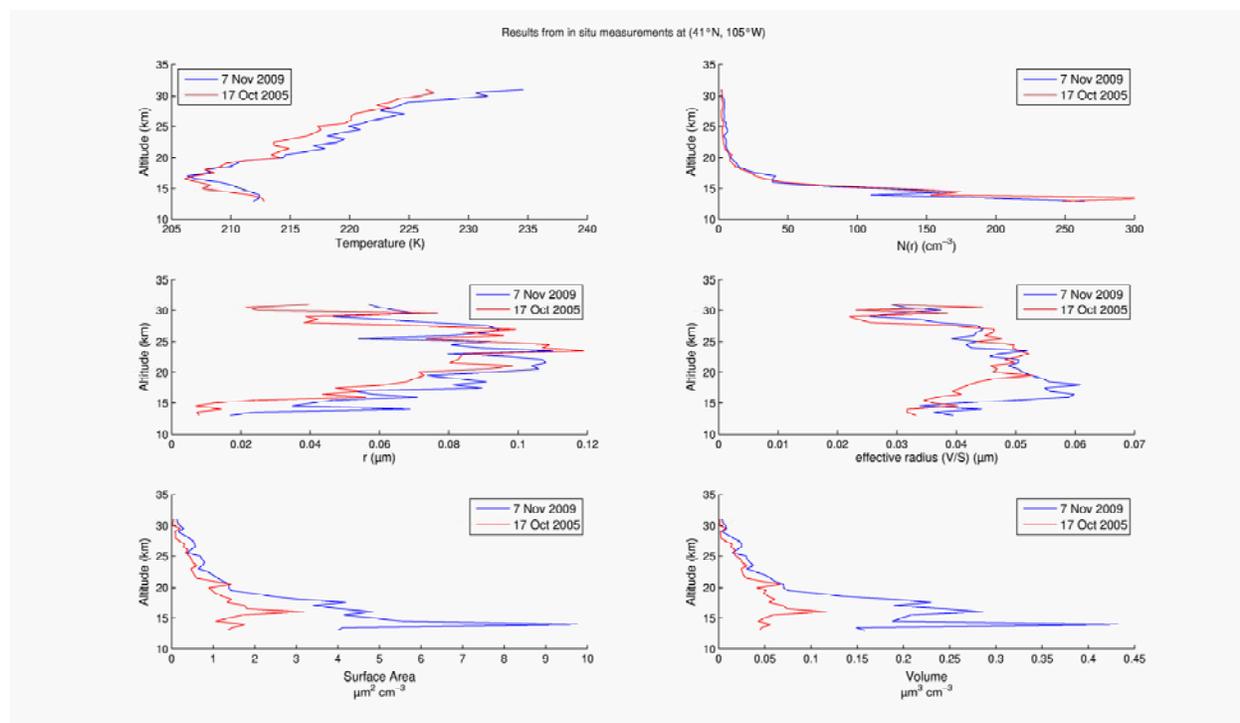
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950 **Figure 7.** OSIRIS retrievals and model output of sulfate aerosol optical depth, as in Figure 5,
951 but scaled to reflect sources of discrepancy. OSIRIS retrievals are unchanged from the values in
952 Figure 5. Model output is scaled using the $\theta = 380$ K line as the tropopause instead of the
953 thermal tropopause. Model output is also multiplied by 0.8 to reflect our overestimation of the
954 initial SO_2 loading, which should have been 1.2 Tg instead of 1.5 Tg. The top and bottom
955 magenta lines denote multiplication of this resultant by 0.78 and 0.38, respectively, to denote
956 changes in optical depth that would result from Ångström exponent scaling. The top line, a
957 multiplication by 0.78, assumes a dry radius of $0.25 \mu\text{m}$, and the bottom line, a multiplication by
958 0.38, assumes a dry radius of $0.08 \mu\text{m}$. All multiplication is performed before taking \log_{10} .



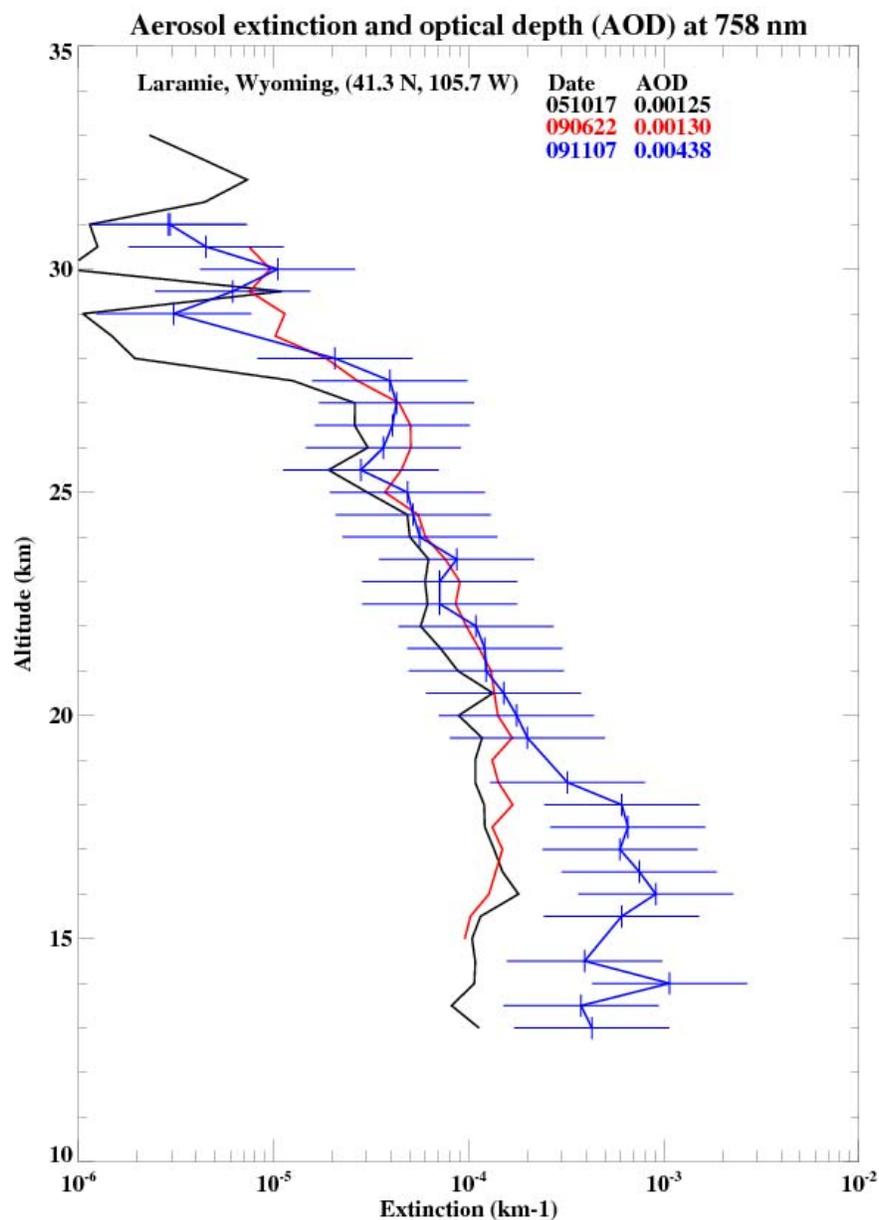
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Figure 8. Profiles of temperature, total aerosol concentration (condensation nuclei), aerosol median radius, effective radius, surface area, and volume derived from size resolved particle concentration measurements from balloon flights from Laramie, Wyoming. Temperature and number concentration are measured, and the other products are derived. The blue line shows measurements on 22 June 2009, ten days after the largest eruption of Sarychev. The red line shows 3 July 2007, which was free of volcanic activity after Soufriere Hills in 2006 and prior to Kasatochi. Measurements are shown from the 22 June 2009 tropopause at 15.0 km to balloon burst at 30.5 km.



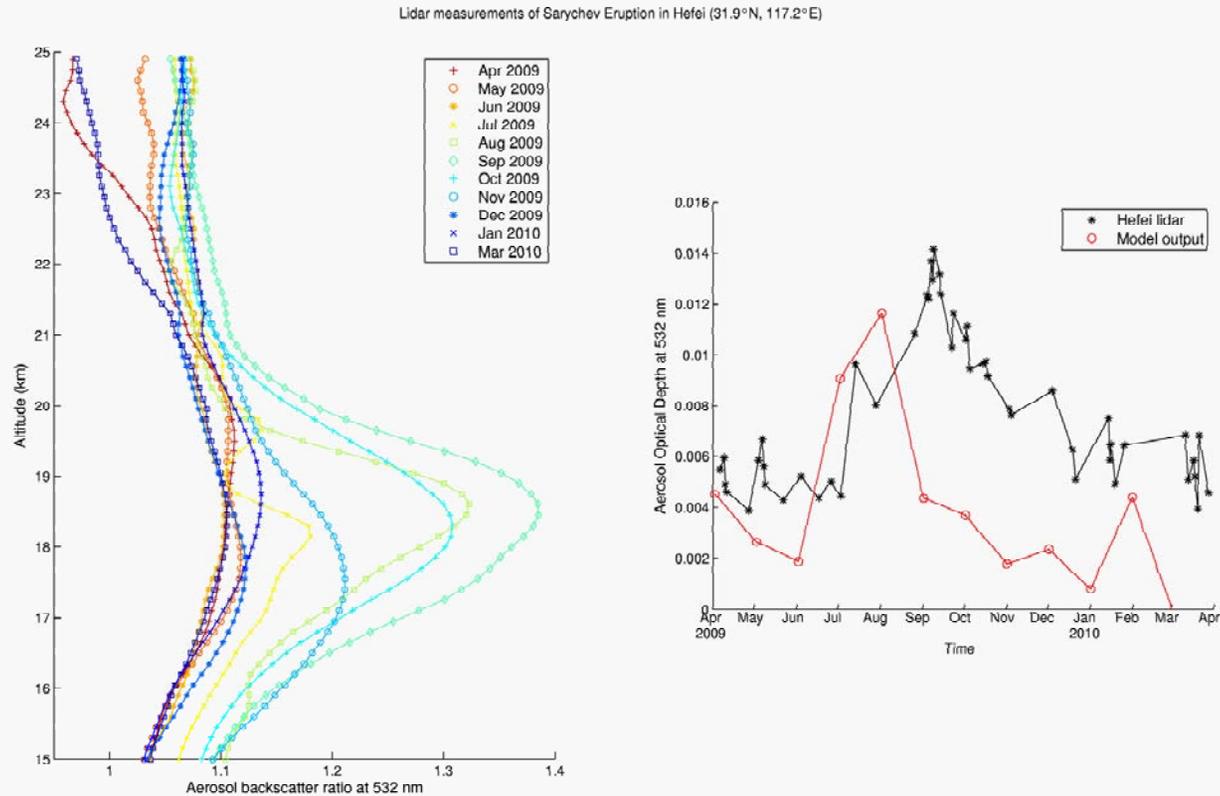
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971 **Figure 9.** Same as Figure 8 but for measurements from Laramie, Wyoming, on 7 Nov 2009
 972 (blue line), several months after the eruption of Sarychev, and 17 Oct 2005 (red line), which was
 973 a period of low perturbations of stratospheric aerosol with otherwise similar atmospheric
 974 conditions to the 2009 measurement. Measurements are shown from the tropopause, at 13.0 km,
 975 to balloon burst at 31.0 km. The volcanic layer appears to have begun to settle through the lower
 976 stratosphere, with a large peak at 14.0 km in altitude.



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Figure 10. Aerosol extinction profiles from in situ measurements on 17 October 2005, 22 June 2009, and 7 November 2009, calculated at 758 nm. The lower limits of the lines are defined by the tropopause on each day. The error bars on 7 November 2009 represent a $\pm 40\%$ uncertainty and apply to the other two profiles as well. The aerosol optical depth (AOD) for each day is shown at the top.

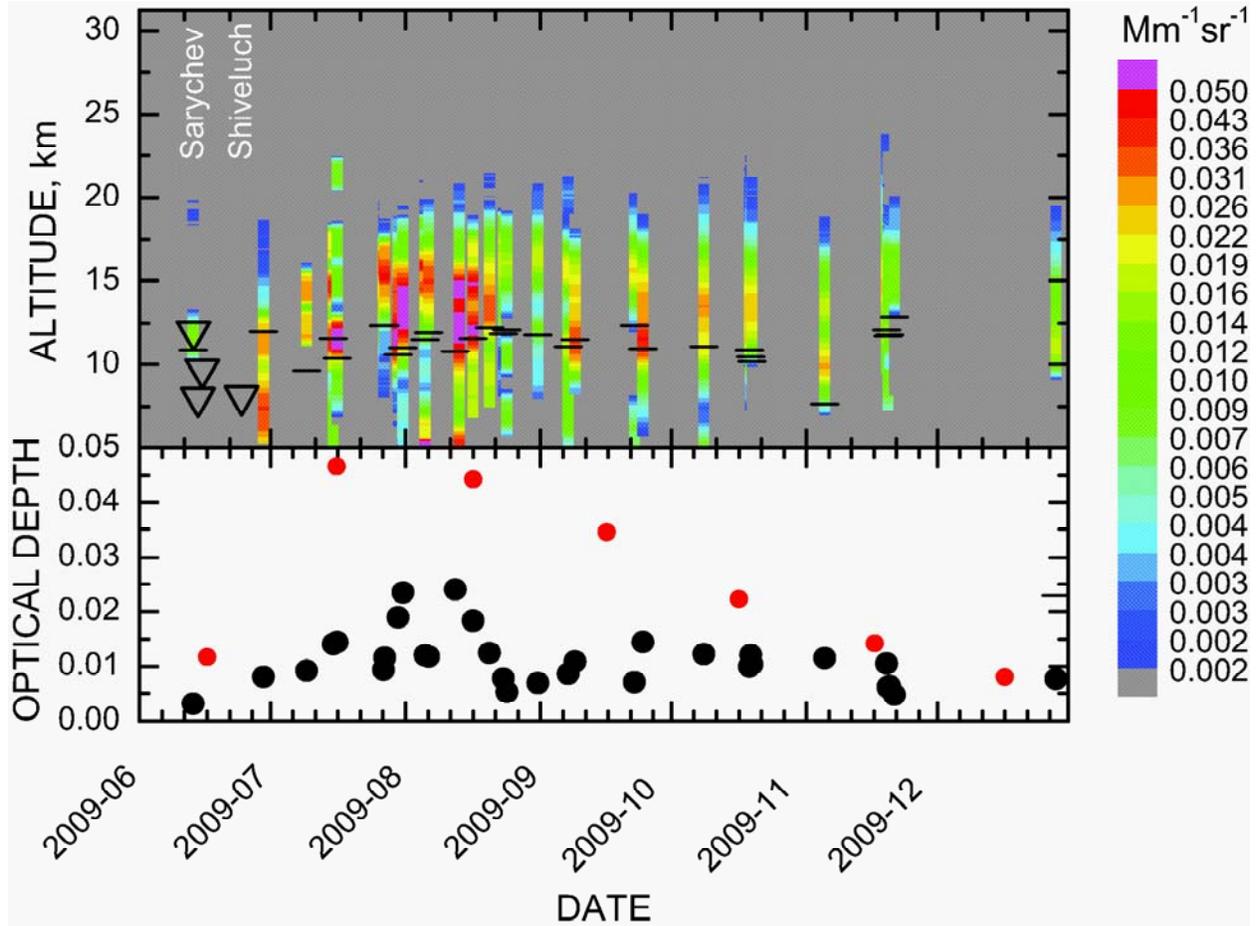


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986 **Figure 11.** Lidar retrievals from Hefei, China compared with ModelE output. The lidar is capable of measuring backscatter up to 25
987 km in altitude. The left panel shows monthly averages of backscatter as a function of altitude, with a maximum in September, 2009.

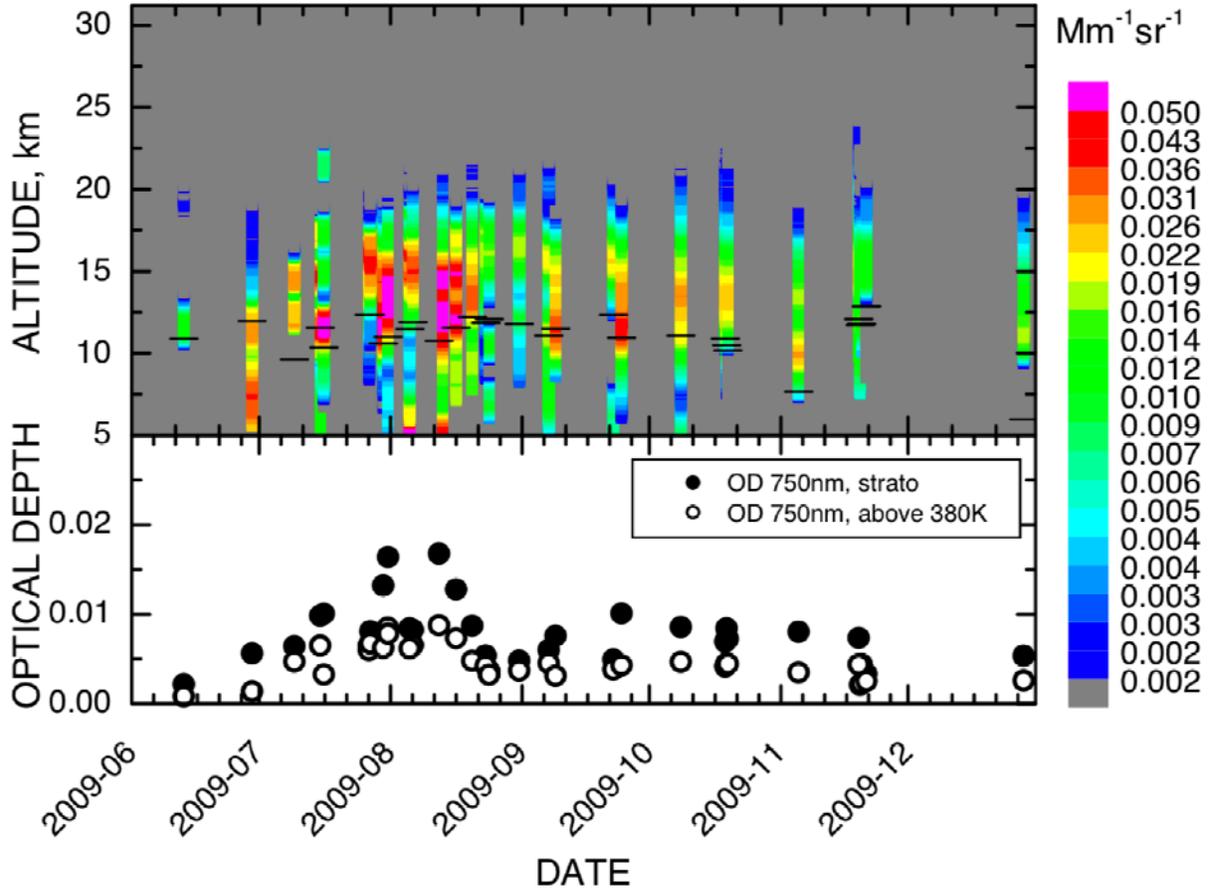
988 The backscatter ratio is defined as the fraction $\frac{\beta_{\text{molecules}} + \beta_{\text{particles}}}{\beta_{\text{molecules}}}$, where β is backscatter, so any values less than 1 are spurious and are

989 likely due to instrument noise. In the right panel, the black line shows integrated (15-25 km) optical depth through the stratosphere,
990 assuming a lidar ratio of 50 sr. The red line shows zonally averaged stratospheric aerosol optical depth calculated by the model in the
991 grid latitude band containing the Hefei lidar (28-32°N). Aerosol concentrations return to background levels by Spring of the year
992 following the eruption.



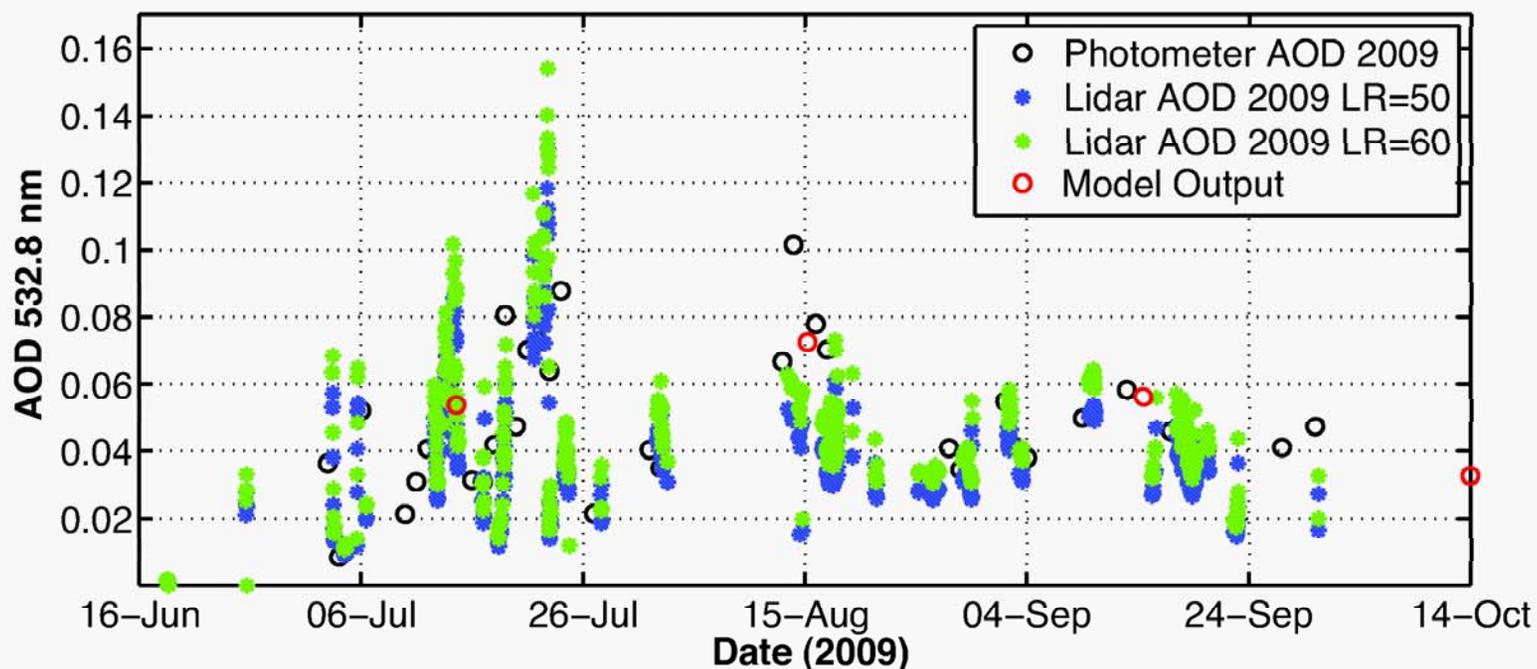
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Figure 12. Backscatter coefficient profiles at 1064 nm and aerosol optical depth at 532 nm from the lidar in Leipzig. Backscatter coefficients are defined as the scattering coefficient (units m^{-1}) at 180 degrees (units sr^{-1}) and are scaled by 10^{-6} , giving units of $\text{Mm}^{-1} \text{sr}^{-1}$. Each strip of backscatter measurements is a 10 day mean profile. Aerosol optical depth was calculated using a lidar ratio of 38 sr, which is the mean value of all cases for which the lidar ratio could be measured. Black dots are stratospheric optical depth measurements calculated using this ratio. Red dots show zonally averaged stratospheric aerosol optical depth calculated by the model in the grid latitude band containing the Leipzig lidar ($48\text{-}52^\circ\text{N}$). Black horizontal lines indicate the height of the tropopause. Triangles show the plume top heights of individual eruptive events. Peak backscatter and optical depth occur in mid-August, and aerosols have returned to low levels by winter following the eruption.



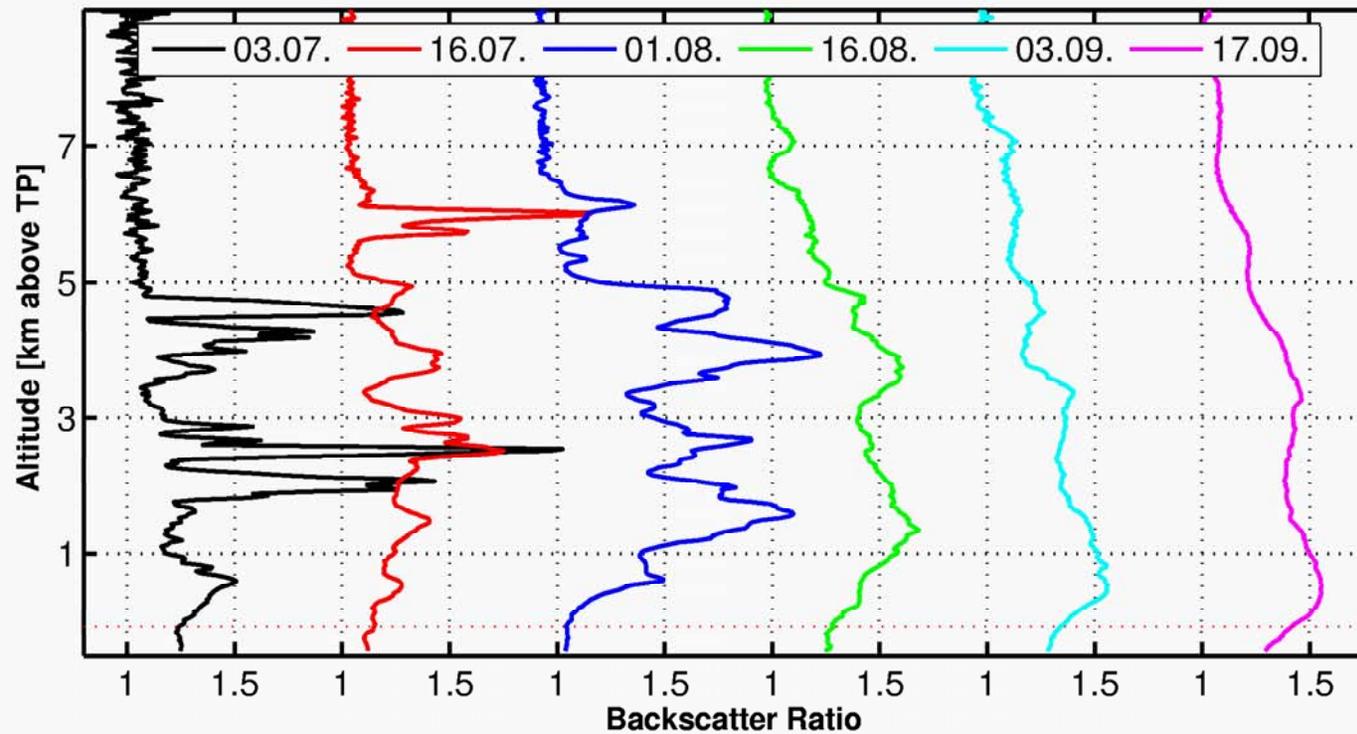
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Figure 13. Same as Figure 12, but optical depth is recalculated at 750 nm, using both the thermal tropopause and the 380 K potential temperature line as the lower bound for integration.



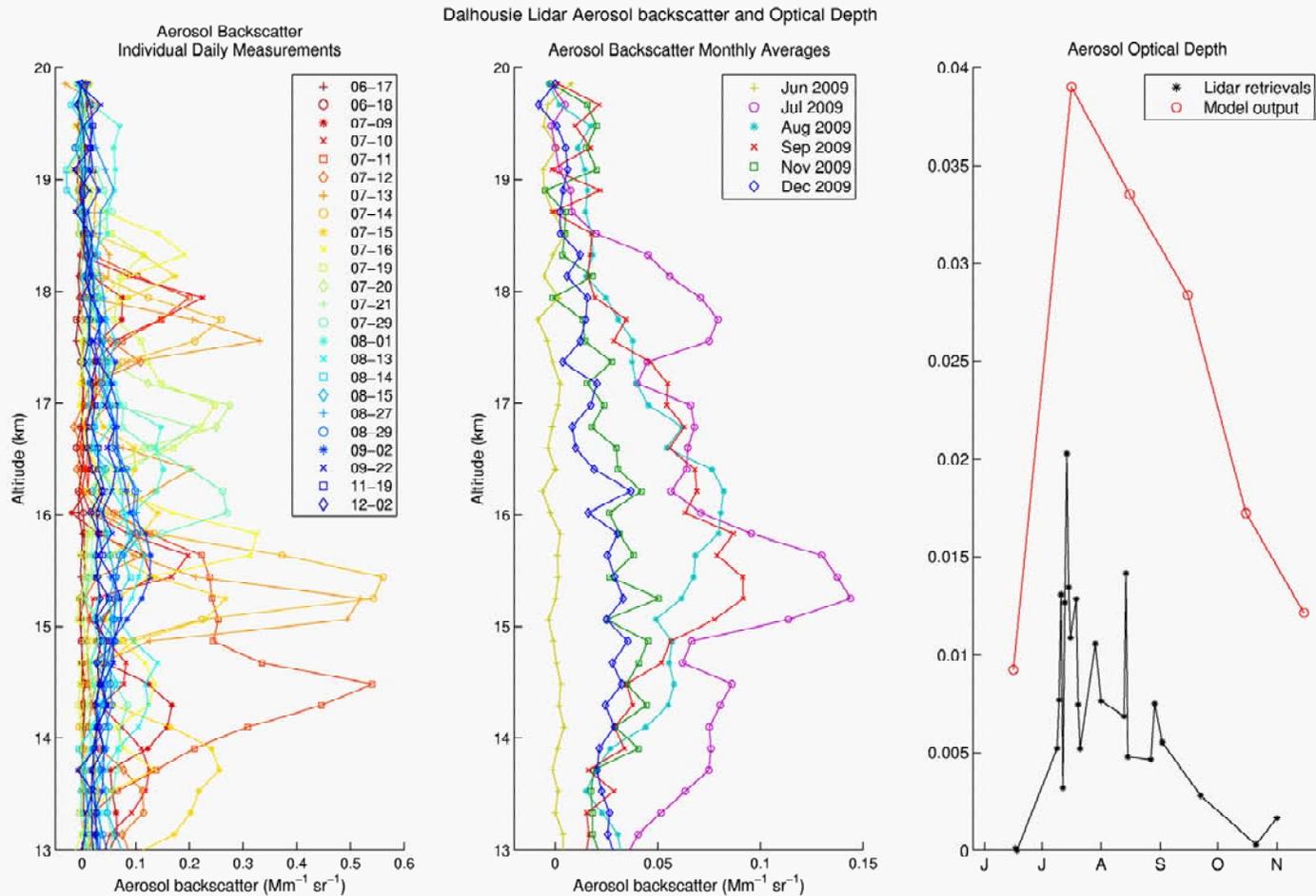
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1013 **Figure 14.** Aerosol optical depth at 532 nm from the KARL lidar and an SP1A sun photometer in Ny-Ålesund, Svalbard. Lidar
 1014 aerosol optical depth was calculated using two different lidar ratios of 50 and 60 sr and integrating the extinction coefficient between
 1015 the thermal tropopause height and 20 km. The lidar ratios were obtained in case studies from 13 July (50±10 sr) and 3 September
 1016 (60±10 sr) according to the transmittance method [Chen *et al.*, 2002]. The tropopause height was derived from co-located daily
 1017 balloon soundings. Photometer AOD are daily means, which are reduced by the monthly long-term means from 1995-2008 without
 1018 extreme events (June: 0.07, July: 0.05, August: 0.045, September: 0.035). Model output is zonally averaged stratospheric aerosol
 1019 optical depth in the grid latitude band containing the Svalbard lidar (76-80°N). Model output values represent monthly averages, so
 1020 they are placed on or near the 15th of each month.



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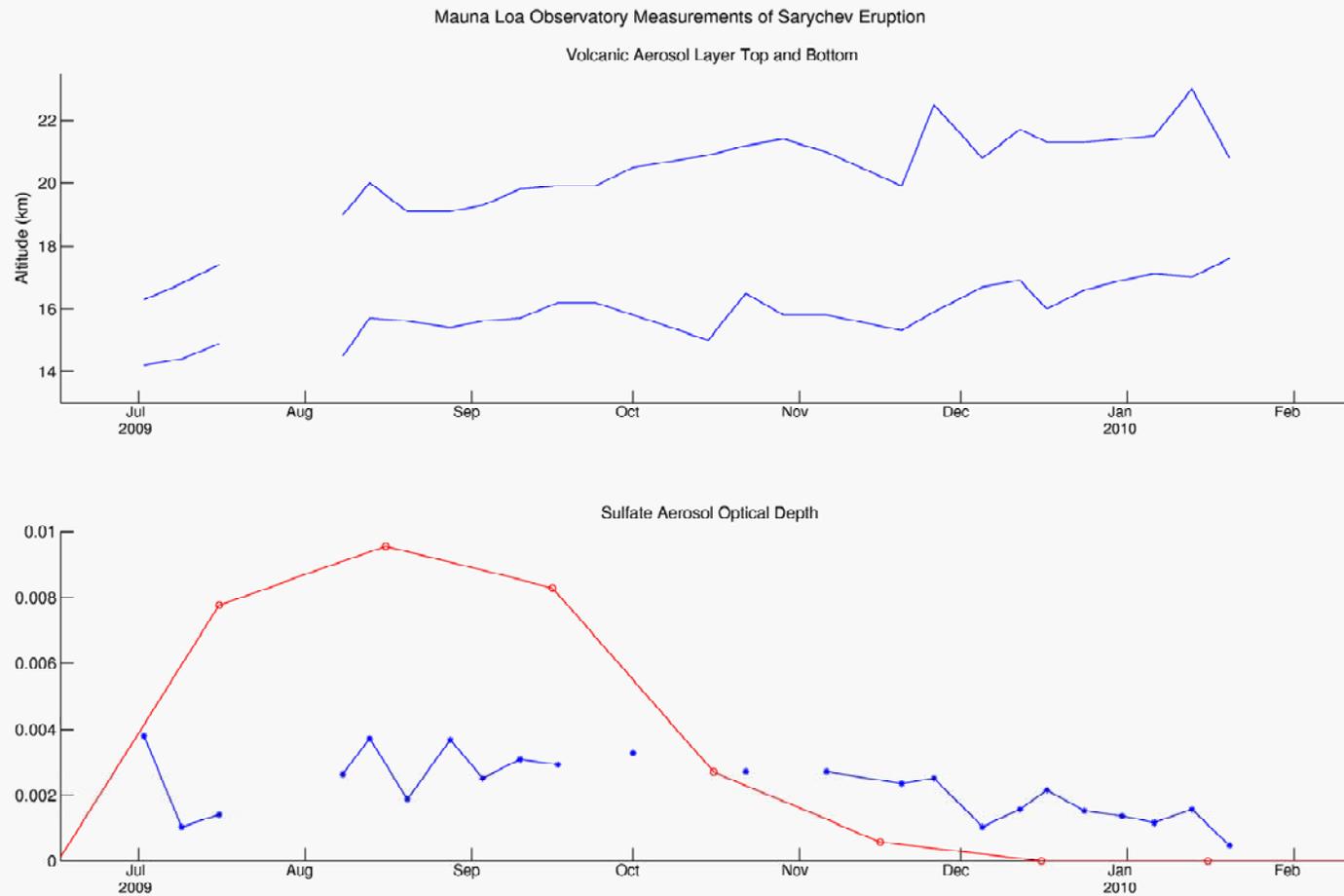
Figure 15. Backscatter ratio profiles at 532 nm for selected days (30 min temporal and 30 m spatial resolution) for the KARL lidar in Ny-Ålesund, Svalbard. Altitude is scaled relative to the thermal tropopause height, which is obtained from co-located daily balloon soundings. In the first two months after the eruption, distinct layers with maximum backscatter ratio above 2 are measured. Late August and September show much smoother profiles with still large values of up to 1.5.



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1029 **Figure 16.** Backscatter and aerosol optical depth from the lidar in Halifax. Backscatter is measured at 532 nm, and the units are the
1030 same as in Figure 12. Measurements below 13 km in altitude show strong interference from cirrus clouds and are omitted. Aerosol
1031 optical depth was calculated using a lidar ratio of 40 sr. Lidar optical depth values are averaged between 15 and 20 km to avoid
1032 interference from cirrus clouds. Red asterisks show zonally averaged stratospheric aerosol optical depth calculated by the model in
1033 the grid latitude band containing the Halifax lidar (44-48°N).

KRAVITZ ET AL.: SARYCHEV OPTICAL DEPTH



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1040

Figure 17. Observations of the Sarychev eruption cloud from the Mauna Loa Observatory. Some observations are missing due to interference from cirrus clouds. The top panel shows the top and bottom of the Sarychev aerosol layer as measured at the Mauna Loa Observatory. In the bottom panel, the blue line shows optical depth calculations from the observatory, which are obtained from measured backscatter using a lidar ratio of 40 sr. The red line shows optical depth as calculated by the model, zonally averaged over the grid band spanning latitudes 16-20°N.