

<sup>1</sup> **Desert dust aerosol age characterized by Mass-Age**  
<sup>2</sup> **Tracking (MAT) of tracers**

Qin Han and Charles S. Zender

<sup>3</sup> Department of Earth System Science, University of California at Irvine

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Q. Han and C. S. Zender, Department of Earth System Science, University of California, Irvine,  
CA 92697-3100. (zender@uci.edu)

4 **Abstract.** We introduce and apply to dust aerosols an efficient method  
5 to track tracer age (time since emission) as a function of space and time in  
6 large-scale geophysical models. Our Mass-Age Tracking (MAT) method fol-  
7 lows the full tracer lifecycles directly and does not depend on proxy, ensem-  
8 ble, or Green’s function techniques. **MAT sends a mass-age tracer through**  
9 **the same algorithms that the host models use to predict tracer mass**  
10 **processes, then estimates age as the ratio of mass-age to mass.** We  
11 apply MAT to size-resolved dust aerosol tracers to study the age of dust that  
12 remains in the atmosphere and the age of dust at deposition. The results in-  
13 clude the first global distribution maps of aerosol age. Dust age varies with  
14 location, time, and particle size and is strongly sensitive to climate—wind  
15 and precipitation in particular. The global average age of dust at deposition  
16 agrees with residence time at  $\sim 2.7$  days while dust in the atmosphere is, on  
17 average, twice as old. As expected, older dust prevails far from sources, at  
18 higher altitudes and in smaller sizes. Dust age exhibits a seasonal cycle, stronger  
19 for larger dust particles, that peaks in April–June, the period of maximum  
20 Asian and North African emissions. The oldest dust at deposition falls in the  
21 Antarctic and South Pacific Convergence Zone about one month after emis-  
22 sion. The mass-weighted ages provided by MAT are useful for investigating  
23 and parameterizing the evolution of aerosol physical and chemical proper-  
24 ties.

## 1. Introduction

25 Atmospheric aerosols have large impacts on climate, biogeochemistry and human health  
26 and these impacts often depend on the aerosol age, i.e., time since emission/formation.  
27 From the moment an aerosol forms or is emitted until its deposition, its composition,  
28 phase, and even shape may change to maintain thermodynamic and chemical equilibrium  
29 with the environment. This “aging” can modify the aerosol scattering and absorption  
30 properties which determine its direct radiative effects, i.e., perturbations of the solar and  
31 terrestrial radiation fields, as well as its indirect radiative effects through influencing cloud  
32 properties. **Aerosol solubility is a crucial factor in atmospheric delivery of bio-**  
33 **available ocean nutrients and micro-nutrients (e.g., Fe) to which ocean biogeo-**  
34 **chemistry is sensitive** [*Krishnamurthy et al., 2009*]. Solubility depends on at-  
35 mospheric processes (trace gas exposure, heterogeneous and photo-chemistry,  
36 cloud processing) during transport [*Spokes and Jickells, 1996; Hand et al.,*  
37 *2004; Luo et al., 2005*]. Age-based parameterization of solubility can be esti-  
38 mated from field measurements with the aid of back-trajectory models [*Hand*  
39 *et al., 2004; Buck et al., 2008*], and from models that represent solubility as  
40 a diffusive or surface-area-controlled process with known time constants [Han  
41 et al., manuscript in prep.]. Knowing the duration of aerosol transport, i.e.,  
42 the aerosol age, can help us understand and thereby better predict or pa-  
43 rameterize the age-related change in aerosol physical and chemical properties  
44 [e.g., *Hand et al., 2004; Luo et al., 2008*]. Here we introduce and apply to

45 **dust aerosols an efficient method to track tracer age in large scale geophysical**  
46 **models.**

47 Aerosol age differs from aerosol lifetime (residence time). **The former is the time**  
48 **since emission/formation, while the latter is the average time that aerosols**  
49 **remain in the atmosphere and is defined as the mean atmospheric burden**  
50 **divided by the total mean source or sink [Prather, 2007]. Age and residence**  
51 **time are closely related in that the global mean mass-weighted aerosol age**  
52 **at deposition asymptotes to the residence time as aerosol sources and sinks**  
53 **approach equilibrium.** Many studies have estimated the residence times of various  
54 aerosols using this definition. Global-mean aerosol residence times vary from just a few  
55 hours for large ( $> 5 \mu\text{m}$  diameter) aerosols such as mineral dust, to more than two  
56 weeks for sub-micron fine aerosols [e.g., Zender *et al.*, 2003; Mahowald *et al.*, 2006a].  
57 Aerosol residence time can also be estimated from the decay of radioactive tracers both  
58 measured [Kuroda *et al.*, 1962; Poet *et al.*, 1972] and in models [Seinfeld and Pandis,  
59 2006]. However, global mean residence times conceal the great spatio-temporal variations  
60 of atmospheric lifetime that arise from local variations in meteorology and deposition  
61 processes (gravitational, turbulent, and wet deposition, i.e., washout). The time since  
62 emission of aerosols in a given air parcel is more relevant to their instantaneous radiative  
63 and chemical influence than is their expected residence time.

64 The age of non-radioactive tracers is difficult to measure and to calculate because it  
65 depends on the history of an air parcel (or mixture of air parcels) rather than on a global  
66 mean budget. Unlike gaseous tracers (e.g., ozone, CO), aerosols experience net motion

67 (e.g., gravitational settling) relative to their air parcel of origin and this complicates their  
68 age determination.

69 Previous studies have estimated tracer age in different ways including the use of age  
70 proxies. One early approach estimated the “photochemical age” of an air mass from  
71 the ratio of hydrocarbons [*Roberts et al.*, 1984]. This approach neglects reactions with  
72 oxidants other than OH so that the “photochemical age” relies on the OH concentrations  
73 and does not change when the OH concentrations are low. Other assumptions—including  
74 that the hydrocarbons used have the same sources, that the sources have a constant  
75 compound composition and that the background concentrations are negligible—also make  
76 this method inaccurate [*Roberts et al.*, 1984; *Parrish et al.*, 1993; *McKeen and Liu*, 1993].  
77 A newer proxy-based approach relies on the differing reaction rates of various stable  
78 isotopic counterparts of the same hydrocarbon, rather than the reaction rates from two  
79 hydrocarbons and OH [*Rudolph and Czuba*, 2000]. This method makes fewer assumptions  
80 and can estimate the average age of each hydrocarbon. However, both of these proxy-  
81 based approaches rely on hydrocarbon reaction rates and neither of them applies well to  
82 other aerosol species.

83 **Previous models have implemented “Age tracer” to track the time since a**  
84 **given seawater parcel was last exposed to the atmosphere, aka the ventilation**  
85 **age [e.g., *Thiele and Sarmiento*, 1990; *England*, 1995]. This method con-**  
86 **structed the continuity equations for age tracers and the age is incremented**  
87 **by one time step during each time step. Similar method has been used to**  
88 **track the age of air [e.g., *Neu and Plumb*, 1999]. It is possible to apply this**

89 **method to tracers. However, each tracer requires its own continuity equation,**  
90 **so this method is not readily generalizable.**

91 More recent research uses additional tracers in trajectory models to track aerosol age  
92 distributions [e.g., *Kleinman et al.*, 2003; *Stohl et al.*, 2003]. **Tracers are tagged to**  
93 **track the emission time and have the same chemical reaction rates as the**  
94 **aerosols that they track [*Kleinman et al.*, 2003]. This approach neglects the**  
95 **mixing of air masses with different ages and looks at only the age of aerosols**  
96 **emitted into the air parcel during the modeled period. Another approach with**  
97 **a Lagrangian trajectory model estimates CO age distribution by tracking the**  
98 **transport time and the contribution from source grids. Though backward**  
99 **simulations yield a higher space and time resolution than the correspond-**  
100 **ing forward simulations, the age distributions calculated by parcel trajectory**  
101 **methods are typically limited by (i) simplified chemistry, (ii) simulation of**  
102 **one pathway per parcel (which can be ameliorated by ensemble techniques as**  
103 **discussed below).**

104 The preceding age-tracking methods have drawbacks in that they (i) rely on partic-  
105 ular emission compositions, (ii) are not easily extensible to other aerosol species, and  
106 (iii) do not account for the mixing of air masses with different ages. General Circulation  
107 Models (GCMs) and/or Chemical Transport Models (CTMs) with more complete aerosol  
108 schemes can solve most of these problems by utilizing the details of transport and deposi-  
109 tion processes. *Krinner and Genthon* [2003] used idealized radioactive tracers to analyze  
110 tracer age at any given place in an atmospheric GCM. Radioactive decay was used as  
111 the only sink, and was intended to implicitly approximate the net removal timescale by

112 all processes. However, the aerosol ages obtained depend on the lifetime assumed for the  
113 radioactive tracer whereas the aerosol ages should be independent of the radioactive tracer  
114 used.

115 The age distribution of an aerosol population is best described by a Probability Density  
116 Function (PDF). Ensemble and Green's Function methods can be used separately and  
117 together to determine the PDF of aerosol ages (and trajectories). *Waugh et al.* [2003]  
118 used the Transit-time PDF (TTPDF) method [*Hall and Plumb*, 1994; *Holzer and Hall*,  
119 2000] to examine the ages of different tracers as well as the temporal variations in tracer  
120 ages. *Holzer et al.* [2003] applied the same method to a CTM to analyse springtime  
121 trans-Pacific atmospheric transport from east Asia. *Primeau and Holzer* [2006] used a  
122 similar technique to track the tracer-independent ventilation rates and the global ocean  
123 age inventory. The TTPDF method yields the transit-time-distributions within a grid  
124 box and thus calculates mean tracer ages accurately. However, this method requires a  
125 large ensemble of simulations when tracking age in time-varying flow fields.

126 *Wagstrom and Pandis* [2009] used the Particulate Matter Source Apportionment Tech-  
127 nology (PSAT) to track the aerosol age in a CTM. The emissions were grouped into bins  
128 based on the time of emission. Each of these emission time periods (ETPs) was treated  
129 as a source category by PSAT. Then the average age of any aerosol species at a particu-  
130 lar location and time was estimated using the mass contributions from each bin and the  
131 average age of each bin. This method is good for estimating average age of aerosols in a  
132 short time period. Though the ETP has only a mild impact on the age estimation, the  
133 number of the ETPs could be very large for long-term runs so that the computational  
134 time increases unreasonably.

135 Our motivation for developing MAT stems from our efforts to model the effects of atmo-  
136 spheric aerosols on ocean biogeochemistry [*Han et al.*, 2008; *Krishnamurthy et al.*, 2009].  
137 For this purpose, we needed an age-tracking method to satisfy the following requirements  
138 that, collectively, are not met by any of the previous methods: firstly, it runs on-line  
139 in GCMs/CTMs so that instantaneous (rather than climatological) tracer age is always  
140 known and can be coupled between atmosphere and ocean; secondly, it is computationally  
141 inexpensive; thirdly, it is generic and applies to any tracer simulated; fourthly, it gives  
142 mean mass-weighted ages (which may be empirically related to aerosol solubility); lastly,  
143 it is deterministic and reproduces the same ages for any given meteorology and tracer  
144 physics.

145 Our Mass-Age Tracking (MAT) method satisfies these requirements. **It requires one**  
146 **additional tracer, for mass-age, per tracer species, and yields results online,**  
147 **during a single simulation rather than requiring post-processing of an ensem-**  
148 **ble of simulations.** Though the MAT method cannot compute the age PDFs for each  
149 grid cell, it estimates the mass-weighted mean aerosol ages accurately and is computa-  
150 tionally efficient. In this work, we apply MAT to investigate the age distributions in the  
151 atmosphere and at deposition of four sizes of dust aerosols.

## 2. Method

152 The MAT method is generic and may be implemented in any geophysical model with  
153 grid-based (Eulerian) representation of mass-conserving tracer physics and dynamics. In  
154 this work we apply MAT to wind generated desert dust, motivated by questions of atmo-  
155 spheric nutrient deposition to understand the age and solubility of aerosol at deposition  
156 [*Spokes and Jickells*, 1996; *Hand et al.*, 2004]. The host GCM is the National Center



157 for Atmospheric Research (NCAR) Community Atmosphere Model, version 3 (CAM3)  
 158 [*Collins et al.*, 2004], configured as in *Flanner et al.* [2007].

159 The dust source, transport and deposition mechanisms follow the Dust Entrainment and  
 160 Deposition Module (DEAD) [*Zender et al.*, 2003]. Dust is entrained into the atmosphere  
 161 through wind mobilization and removed by dry gravitational settling, turbulent dry depo-  
 162 sition and wet deposition during precipitation events. Between the sources and sinks, dust  
 163 is advected and diffused as a passive tracer by the transport processes used in CAM3, in-  
 164 cluding vertical diffusion, shallow convection, deep convection and semi-Lagrangian tracer  
 165 transport. **Dust is divided into four size bins based on the diameters of the parti-**  
 166 **cles (Table 1) and particle size distributions are assumed to be time-invariant**  
 167 **within each bin. A dust particle does not change its effective size or mass**  
 168 **through its lifecycle and there is no exchange between bins.**

The implementation of the MAT method is straightforward. **For each tracer species**  
**in the host model (e.g., GCM or CTM), MAT prognoses one additional tracer:**  
**the mass-age (the product of aerosol mass and age, in units of [kg sec]) of the**  
**tracer. In this case, the mass-age of each dust aerosol size class is carried**  
**through each lifecycle process mentioned above at all model grid points and**  
**the mass-weighted age ( $A$ ) of dust aerosols is then derived by dividing mass-**  
**age ( $mA$ ) by mass ( $m$ ). If we describe the change of dust dry mass in the**  
**model as:**

$$\frac{dm}{dt} = \mathcal{L}(m) + S \quad (1)$$

then the change of dust mass-age can be described as:

$$\frac{dmA}{dt} = \mathcal{L}(mA) + m \times T \quad (2)$$

169 where  $S$  is the dust source from wind mobilization,  $T$  is the length of time for  
 170 one model time step (20 minutes in our case), and the operator  $\mathcal{L}$  denotes all  
 171 other dust lifecycle processes including vertical diffusion, shallow convection,  
 172 deep convection, semi-Lagrangian tracer transport, and dry and wet deposi-  
 173 tion. Our method is very similar to traditional age tracer methods [*Thiele*  
 174 *and Sarmiento, 1990; England, 1995; Neu and Plumb, 1999*]. However, the  
 175 elegance of MAT is that the lifecycle operator  $\mathcal{L}$  is the same for mass and  
 176 mass-age, so that the same algorithms and codes used on tracer mass can be  
 177 reused on mass-age. There are exceptions, discussed below, for algorithms  
 178 that contain mass-specific switches. Both dust mass ( $m$ ) and mass-age ( $mA$ )  
 179 are zero initially and they also have the same sinks (wet and dry deposition).  
 180 The only difference is that the source of dust mass is emission, while the source  
 181 of mass-age is the internal increment of one time step per unit mass during  
 182 each time step. Note that we use dust to demonstrate the use of MAT for  
 183 an inert tracer. MAT applies to other tracers, even to chemically and ther-  
 184 modynamically active species, as long as all the processes are included in the  
 185 operator  $\mathcal{L}$ .

186 If the host model conserves tracer mass then it will conserve tracer mass-age  
 187 too, either automatically, or after some coaxing described below. In equilib-  
 188 rium, the global total mass-age deposited in a period equals the total mass-age  
 189 added in the same period. The former is the product of global mean mass-

190 weighted age of dust at deposition and the mass of dust deposited in this  
191 period. The latter is the product of the mass of dust that remains in the  
192 atmosphere and the length of this period. Thus, by definition, the global av-  
193 erage mass-weighted age of deposited dust equals the global dust residence  
194 time at equilibrium.

195 We uncovered one obstacle to accurate implementation of MAT by verifying  
196 the conservation of the mass-age tracer. To wit, dynamic and physical process  
197 algorithms (e.g., advection, wet deposition) often contain non-linear switches  
198 or conditions (if-then statements) that depend on the tracer value being in the  
199 expected range. Such conditions might be that tracer mixing ratio and mass  
200 should not be negative, or that tracer mixing ratio should exceed some small  
201 value  $\epsilon$  ( $\epsilon \sim 10^{-30}$ ) so that floating point arithmetic will not underflow in single  
202 precision. Unpredictable results arise when these algorithms encounter the  
203 mass-age tracer since the same numerical limit has different effects on mass  
204 than on mass-age. We solve this problem by making parallel approximations  
205 to the mass-age tracer as to mass. Thus the equivalent global mean mass-  
206 age threshold  $\epsilon_{mA}$  will be  $\epsilon$  multiplied by the factor  $A$  (e.g.,  $A \sim 3 \times 10^7$  for a  
207 one year old tracer). Fortunately these problems usually occur only in “corner  
208 cases” such as extremely low mass concentrations where the simple parallel  
209 approximations are acceptable because they affect negligible mass and mass-  
210 age. Hence in our implementation of MAT, the mass-age tracers are conserved  
211 to the same precision as the mass tracers.

212 Here we use MAT to track the mass-weighted age of dust that remains in the atmosphere  
213 **(for conciseness, referred to hereafter as dust aloft)**, and of dust deposited to  
214 the surface as functions of space, time, and dust size. The results shown below are  
215 averages and time series from the last ten years of 20-year equilibrium present day climate  
216 simulations.

### 3. Results: dust age in present-day climate

#### 3.1. Temporal evolution of dust age and residence time

217 Dust lifetime, which as mentioned above, is also known as its residence time, is the  
218 average time that dust particles are expected to stay in the atmosphere and is defined as  
219 the total global dust burden in the atmosphere divided by the dust deposition (or mobi-  
220 lization) rate. Dust age is defined as the time elapsed since a dust particle entered the  
221 atmosphere and is computed by MAT. Average dust residence times range from 1.5 to 8.2  
222 days from the largest dust sizes modeled (5–10  $\mu\text{m}$  diameter) to the smallest sub-micron  
223 dust sizes (Table 1). The residence times compare well with *Mahowald et al.* [2006b] who  
224 also used the CAM GCM. Residence times computed from the dust field simulated by the  
225 MATCH CTM and driven by National Center for Environment Prediction (NCEP) ana-  
226 lyzed meteorology for the period 1990–1999 [*Zender et al.*, 2003] differ from the residence  
227 times calculated from CAM. The long-term average residence time in MATCH is more  
228 than twice the CAM residence time for bin 1 and is  $\sim 3$  days longer for bin 2. For bins  
229 3 and 4, the MATCH residence times are slightly shorter than those from CAM. These  
230 differences arise because MATCH and CAM have slightly differing dust entrainment and  
231 deposition schemes, and strongly differing meteorologies, in particular wind and precipi-  
232 tation. The residence times for small dust particles are more sensitive to the meteorology

233 than those for large dust particles, because smaller dust particles are more susceptible to  
234 long range transport and wet scavenging. MATCH and CAM utilize different horizontal  
235 resolutions which would also produce differing dust fields [*Zender et al.*, 2004].

236 The ages of dust aloft and of dust at deposition both decrease as the particle size in-  
237 creases. The MAT-calculated global average ages of deposited dust and the traditional  
238 residence times are not identical but agree as well as expected (Table 1, Figure 1). The  
239 small differences between them reflect the ever changing atmospheric dust burden. Ap-  
240 proximations made in CAM to conserve mass in a positive-definite manner [*Collins et al.*,  
241 2004] and the parallel approximations we make to conserve mass-age could also contribute  
242 to these differences. Dust aloft is usually 1–2 days older than dust at deposition except  
243 for size bin 2 (1–2.5  $\mu\text{m}$  diameter). As discussed below, this is due to size-dependent  
244 deposition processes and ages.

245 The residence times and ages of dust at deposition have strong annual cycles with  
246 longer ages/residence times in Northern Hemisphere (NH) spring/summer (Figure 1).  
247 The seasonal cycles of residence times and ages at deposition of smaller dust have a sharp  
248 drop following the annual maximum. The residence times and ages at deposition of larger  
249 dust drop more slowly and the minima are seen in NH winter. The ages of all sizes of  
250 dust aloft have clear seasonal cycles except the age of sub-micron (size bin 1) dust aloft.  
251 The seasonal cycles of dust ages and residence times are probably caused by the change  
252 of dust plume locations and the precipitation of the plume region. However, it is not clear  
253 why the seasonal patterns are different for different dust sizes.

### 3.2. Spatial distribution of dust age at deposition and aloft

254 The MAT method reveals the mass-weighted ages of dust at deposition globally (Fig-  
255 ure 2a,b). Dust is relatively young when deposited near source regions and is relatively  
256 old when deposited far from source regions. Dust deposited at inland deserts in low and  
257 middle latitudes ranges in age from 1–2 days old (sub-micron dust) to only a few hours  
258 (5–10  $\mu\text{m}$ ). Dust deposited to remote oceans and to polar regions is more than two weeks  
259 old (size bin 1) and more than one week old (size bin 4). Note that the largest dust settles  
260 relatively quickly ( $\sim 300 \text{ m day}^{-1}$ ) so that mass fraction (and mass flux) of this dust that is  
261 older than one week is extremely small. Oceans downwind of dust source regions, such as  
262 the equatorial Atlantic, receive intermediate-aged dust. The Southern Hemisphere (SH)  
263 receives older deposited dust than the NH since the SH is relatively farther from dust  
264 sources. The global average age of dust at deposition is much closer to the NH than the  
265 SH age since the NH deposition rate is 10 times more than the SH rate and we report the  
266 mass-weighted age. Of course in terms of chemical processing and effects on solubility it  
267 is the local not the global mass-weighted ages that matter.

268 The age of dust aloft is also derived by using the mass-age tracers. The dust ages  
269 increase with height and, as expected, modelled dust in the stratosphere is much older  
270 than in the troposphere: more than 2 years for sub-micron dust and 2–3 months for 5–10  
271  $\mu\text{m}$  dust (Figure 3). Note that the CAM GCM employed is primarily a tropospheric GCM  
272 so distributions of tracers in the stratosphere should not be over-interpreted. The ages  
273 of dust near the surface are consistent with the ages of deposited dust. Dust deposited  
274 around 30 °N and 30 °S has very short ages since the dominant dust sources—subtropical

275 deserts—are located there. Polar regions have old dust ages since they are very far from  
 276 dust sources.

### 277 **3.2.1. MAT modified to diagnose spatial trajectories**

278 The oldest dust deposited outside the Antarctic falls in the equatorial Pacific region  
 279 (0–10 °S) along the South Pacific Convergence Zone (SPCZ). Dust falling in this region  
 280 northeast of Australia (Figure 2) can be more than one month old. The atmospheric dust  
 281 aloft at 10 °S is also  $\sim 5$  days older than the surroundings (Figure 3). We examined the  
 282 extent to which dust passing through the stratosphere contributes to the old age of the  
 283 dust in this region. Before we get into this discussion, we caution that very little dust  
 284 falls there. Thus the age of SPCZ dust is of interest for processes which strongly depend  
 285 on the tail of the aerosol age distribution.

286 At least two competing hypotheses could explain the great mass-weighted age of SPCZ  
 287 dust. First, the age may be influenced by dust that entered the upper troposphere or  
 288 lower stratosphere in deep convective events, and exited likewise or through settling. It  
 289 could also be dust that was emitted from North America and Africa and transported by  
 290 slow easterlies in the equatorial boundary layer. We are unaware of any measurement of  
 291 dust aerosol ages in the SPCZ region.

Estimating how much tracer traverses a given spatial region requires only a slight mod-  
 ification of the MAT method. **We track the the mass of dust that has been to the  
 stratosphere ( $mf$ ) by:**

$$\frac{dmf}{dt} = \mathcal{L}(mf) \quad (3)$$

292 **where  $mf$  is initially zero everywhere and then equals the total dust mass ( $m$ )**  
 293 **for any stratospheric region (defined to be above the model interface at 92**

294 hPa since the tropical tropopause is normally below 100 hPa). Here  $mf$  is  
295 conserved once created by dust mass that enters the stratosphere by rising  
296 above the 92 hPa model level. The mean mass-weighted fraction of dust that  
297 has been to the stratosphere ( $f$ ) thus equals the ratio of  $mf$  to  $m$ .

298 Thus the modified MAT estimates the fraction of dust that has ascended,  
299 at least once, above 92 hPa. This is a conservative estimate (lower bound)  
300 of the fraction of dust that has been in the stratosphere (Figure 4a,b) since  
301 the tropopause at higher latitudes is usually much lower than 92 hPa taken  
302 to demarcate the tropical tropopause. We caution that although the cross-  
303 tropopause transport (e.g., deep convection) in CAM has been evaluated [*Rasch et al.*,  
304 2006; *Williamson and Rasch*, 1994], whether the fluxes of aerosols by this transport are  
305 correctly represented remains unclear. Less than 5% of the smallest dust particles and  
306 less than 0.5% of the largest dust particles deposited in the SPCZ traversed the strato-  
307 sphere. We also traced dust that rose above 208 hPa and found that more than 40% of  
308 sub-micron dust and more than 50% of 5–10 micron dust deposited in the SPCZ traversed  
309 the upper troposphere lower stratosphere (UTLS) region between 92 hPa and 208 hPa  
310 (Figure 4c,d). The large fraction of UTLS dust explains well the old mass-weighted ages  
311 of the deposited dust in the SPCZ. Strong and deep convective events can explain why  
312 the SPCZ has the largest fraction of stratosphere-influenced dust. It is unclear why the  
313 intertropical convergence zone (ITCZ) to the north does not follow the same pattern.

### 3.3. Dust age by deposition processes

314 Not only does dust deposited have different ages from dust aloft, dust deposited by  
315 wet and dry deposition have different ages (Table 1). The age of dry-deposited dust



316 reflects the dust age near the surface while the age of wet-deposited dust represents the  
317 mass-average dust age in higher layers where precipitation originates. Since the age of  
318 dust aloft increases with altitude, dust aloft and wet-deposited dust are much older than  
319 dry-deposited dust for all dust size ranges. Dust aloft is slightly older than wet-deposited  
320 dust for sub-micron dust and younger for larger dust. Since dust deposition is dominated  
321 by dry deposition for larger particles (size bin 3 and 4) and by wet deposition for smaller  
322 particles (size bin 1 and 2), the age of deposited dust is determined by the age of dry-  
323 deposited dust for size bins 3 and 4 and by the age of wet-deposited dust for size bins 1  
324 and 2. Thus deposited dust is usually younger than dust aloft except for size bin 2.

325 **The global mass of dust in each size class is binned in days according to**  
326 **age (Figure 5). Although dust ages range from hours to years, most dust (by**  
327 **mass) is younger than two weeks.** 90% of wet-deposited dust is younger than twice  
328 the global average age of wet-deposited dust and 90% of dry-deposited dust is younger  
329 than three times the average dry-deposited dust age. Large dust particles are concentrated  
330 in younger ages while small dust size bins have a relatively flat distribution curve. Dry-  
331 deposited dust is more concentrated in short ages than is wet-deposited dust. The portion  
332 of dust aloft aged less than 20 days is between the two kinds of deposited dust but more  
333 dust aloft is older than 25 days due to the influence of (very old) stratospheric dust.

334 The average age of wet-deposited dust is almost twice that of the dry-deposited dust,  
335 but their age distributions have very similar spatial patterns (Figure 2c-f). Most regions  
336 have deposited dust older than the global average dust age. For dry-deposited dust, only  
337 dust deposited in deserts has age near the global average dust age. Again, since we are

338 calculating the mass-weighted age, the desert regions count for more than half of the  
339 global total dust deposition and thus dominate the global average age of deposited dust.

#### 4. Discussion

340 We developed the Mass-Age Tracking (MAT) method as an efficient and accurate means  
341 of estimating the mass-weighted age of tracers. For this particular purpose, MAT has  
342 many advantages over other experimental and modeling tracer age estimation methods.  
343 MAT does not depend on age proxies and thus tracks mass-weighted tracer ages accurately  
344 at any location and any time. MAT can be applied to any mass-conserving tracer (e.g.,  
345 aerosol, gas, isotope, chemical) in geophysical models. In particular, MAT embeds rather  
346 naturally in GCMs and CTMs, where it may be implemented as a second call to all the  
347 mass-transformative algorithms. Thus MAT can estimate not only the tracer ages in the  
348 current climate, but also in future and past climates.

349 Whereas the TTPDF method [*Holzer and Hall, 2000*] tracks a full spectrum of age PDFs  
350 and is appropriate for studies that need age distributions inside each grid cell, MAT tracks  
351 only one mass-weighted age at one grid cell for each tracer. For each tracer species, MAT  
352 consumes computer time equal to that required to simulate the lifecycle of the tracer. In  
353 our case of tracking four aerosols in a tropospheric GCM, MAT increased total simulation  
354 time by 25%. MAT can also be modified for other uses, e.g., examining the age-influence  
355 of each aerosol lifecycle process, or estimating the fractions of tracers traversing particular  
356 regions.

357 We applied the MAT method to four sizes of dust aerosols ranging from 0.1–10.0  $\mu\text{m}$  in  
358 diameter. The results provide insights into the roles of transport, deposition processes and  
359 meteorology on dust age. Climate, mostly wind and precipitation, have a large impact on

360 dust age, especially for smaller dust particles. The global average dust ages at deposition  
361 range between 1.4–7.8 days in the current climate. In the same climate, dust age varies  
362 with location, time and particle size. Dust that is further from its origin, higher, or  
363 smaller, is likely to be older than otherwise. Larger dust in summer is clearly older than  
364 in winter, but there is no obvious annual pattern for smaller dust. While fine dust lofted  
365 into the stratosphere may remain there for years, over 90% of dust deposits within two  
366 weeks.

367 We presented the first global distribution maps of aerosol age. With the distribution of  
368 aerosol ages, aerosol properties and lifecycles can be further examined, understood and  
369 accounted for. For example, aerosols experience chemical processing during transport  
370 and comparisons between chemical models and observations show that older dust may be  
371 more soluble because it has experienced more or longer exposure to sunlight, clouds, and  
372 heterogeneous chemistry [*Hand et al.*, 2004]. **Thus ocean areas far from dust source**  
373 **regions could receive dust with a solubility that is larger than the global**  
374 **average** and therefore more soluble iron could be delivered to these low dust deposition  
375 regions than estimated by globally uniform solubilities [*Moore et al.*, 2004; *Han et al.*,  
376 2008]. Conversely, oceans immediately downwind of deserts may receive dust that is less  
377 soluble [*Baker et al.*, 2006]. The MAT method may be a useful means of accounting such  
378 age-related consequences in large scale models.

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**Table 1.** Dust ages and residence times (in days) in equilibrium present-day climate

Size Bin	Dust diameter ( $\mu\text{m}$ )	MATCH*		CAM global				CAM Northern Hemisphere		CAM Southern Hemisphere	
		$\tau$	$\tau$	$\alpha_{dps}$	$\alpha_{wet}$	$\alpha_{dry}$	$\alpha_{air}$	$\alpha_{dps}$	$\alpha_{air}$	$\alpha_{dps}$	$\alpha_{air}$
Bin1	0.1-1.0	16.9	7.8 $\pm$ 0.2	7.8 $\pm$ 0.2	7.9 $\pm$ 0.2	3.9 $\pm$ 0.1	8.4 $\pm$ 0.1	7.6 $\pm$ 0.2	7.9 $\pm$ 0.1	9.2 $\pm$ 0.4	12.3 $\pm$ 0.3
Bin2	1.0-2.5	10.9	7.3 $\pm$ 0.2	7.2 $\pm$ 0.2	7.6 $\pm$ 0.2	3.9 $\pm$ 0.2	7.1 $\pm$ 0.1	7.0 $\pm$ 0.2	6.86 $\pm$ 0.09	8.4 $\pm$ 0.4	9.0 $\pm$ 0.3
Bin3	2.5-5.0	3.5	3.8 $\pm$ 0.1	3.7 $\pm$ 0.1	6.1 $\pm$ 0.1	1.82 $\pm$ 0.08	5.40 $\pm$ 0.06	3.6 $\pm$ 0.1	5.30 $\pm$ 0.06	4.5 $\pm$ 0.1	6.2 $\pm$ 0.1
Bin4	5.0-10.0	1.1	1.37 $\pm$ 0.04	1.37 $\pm$ 0.04	3.7 $\pm$ 0.1	0.92 $\pm$ 0.03	3.15 $\pm$ 0.05	1.34 $\pm$ 0.04	3.11 $\pm$ 0.06	1.67 $\pm$ 0.03	3.44 $\pm$ 0.05
All sizes	0.1-10.0	4.3	2.69 $\pm$ 0.06	2.67 $\pm$ 0.07	5.9 $\pm$ 0.2	1.10 $\pm$ 0.04	5.47 $\pm$ 0.08	2.56 $\pm$ 0.07	5.30 $\pm$ 0.07	3.7 $\pm$ 0.2	6.9 $\pm$ 0.2

\* Global mean residence time (in days) from *Zender et al.* [2003].

<sup>484</sup>  $\tau$  is residence time;  $\alpha_{dps}$  is age at deposition;  $\alpha_{wet}$  is age at wet deposition;  $\alpha_{dry}$  is age at dry deposition;  $\alpha_{air}$  is age of dust aloft.

**Figure 1.** Global mass-weighted average ages of dust in the atmosphere (red), ages of dust at deposition (blue) and dust residence times (black) as a function of time (model year 17–20) for four dust sizes. Dashed lines show the average residence times. Grey area denotes April, May and June in each year.

**Figure 2.** Spatial distributions of deposited dust ages (days) averaged through the last 10 model years: (a) ages of total (wet+dry) deposited sub-micron (size bin 1) dust, (b) ages of total deposited 5–10  $\mu\text{m}$  (size bin 4) dust, (c) ages of wet-deposited size bin 1 dust, (d) ages of wet-deposited size bin 4 dust, (e) ages of dry-deposited size bin 1 dust, (f) ages of dry-deposited size bin 4 dust.

**Figure 3.** Vertical and meridional age distributions (days) for four sizes of dust in the atmosphere averaged through the last ten model years.

**Figure 4.** The percentage of deposited dust that has been in (a) above 92.37 hPa for sub-micron (size bin 1) dust, (b) above 92.37 hPa for 5–10  $\mu\text{m}$  (size bin 4) dust, (c) between 92.37–208.15 hPa for sub-micron dust, (c) between 92.37–208.15 hPa for 5–10  $\mu\text{m}$  dust.

**Figure 5.** The age probability distributions of dust. Red for dust in the atmosphere, green for wet-deposited dust and yellow for dry-deposited dust. The crosses denote that 90% of wet or dry deposited dust in mass have ages younger than the ages at the crosses.