Wet deposition in a global size-dependent aerosol transport model: 2. Influence of the scavenging scheme on 210Pb vertical profiles, surface concentrations, and deposition

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Wet deposition in a global size-dependent aerosol transport model

2. Influence of the scavenging scheme on $^{210}$Pb vertical profiles, surface concentrations, and deposition

W. Guelle, Y. J. Balkanski, J. E. Dibb, M. Schulz, and F. Dulac

Abstract. The main atmospheric sink for submicron aerosols is wet removal. Lead $^{210}$, the radioactive decay product of $^{222}$Rn, attaches immediately after being formed to submicron particles. Here we compare the effects of three different wet-scavenging schemes used in global aerosol simulations on the $^{210}$Pb aerosol distribution using an off-line, size-resolved, global atmospheric transport model. We highlight the merits and shortcomings of each scavenging scheme at reproducing available measurements, which include concentrations in surface air and deposition, as well as vertical profiles observed over North America and western and central North Pacific. We show that model-measurement comparison of total deposition does not allow to distinguish between scavenging schemes because compensation effects can hide the differences in their respective scavenging efficiencies. Differences in scavenging parameterization affect the aerosol vertical distribution to a much greater extent than the surface concentration. Zonally averaged concentrations at different altitudes derived from the model vary by more than a factor of 3 according to the scavenging formulation, and only one scheme enables us to reproduce reliably the individual profiles observed. This study shows that ground measurements alone are insufficient to validate a global aerosol transport model.

1. Introduction

Very little has been done to evaluate tropospheric aerosol vertical distribution predicted from models against actual measurements. This represents a clear gap in our ability to adequately represent aerosol mass and number concentrations, to assess their direct radiative effect, and to estimate the heterogeneous reactions that take place at their surfaces. A description of the processes that affect aerosol number concentration necessitates a good representation of the fate of the aerosol in and below clouds.

Boucher [1995, p. 87] highlighted the differences that arise in sulfate distributions between the models MOGUNTIA and IMAGE. Not only are the amplitude of the resulting radiative forcings different, but the timing of its northern hemisphere maximum is shifted by nearly 3 months between the two models. Although these discrepancies are not only due to wet scavenging, the important role it has as the main sink for $\text{SO}_2^-$ and an important pathway for $\text{SO}_2$ removal makes it one of the best candidates to explain these differences. We are limited in terms of validation of the sulfate distribution by the number of vertical profiles that have been measured for both sulfate and $\text{SO}_2$ [Chin et al., 1996].

Heterogeneous chemistry has been documented to take place on the surface of mineral aerosol [Mamane and Gotlieb, 1989] or sea-salt particles [Finlayson-Pitts, 1983]. The main limitations for assessing the impact of these aerosols on tropospheric chemistry are twofold: (1) a better knowledge of the sticking coefficients and (2) an evaluation of the surface of the aerosol available at any given time and place for these reactions to take place. We therefore need not only to represent mass correctly but also to compute aerosol size distribution. Until recently, global aerosol models computed only the aerosol mass. A step toward an accurate computation of the aerosol surface and number concentration was done either using 3 [Genthon, 1992], 8 [Tegen and Lacis, 1996; Gong et al., 1997], 10 [Dentener et al., 1996], or 20 size bins [Schulz et al., 1998], or using a spectral scheme that deals with modal parameters of size distribution [Schulz et al., 1998]. Since heterogeneous chemistry can occur
in the presence of SO$_2$ in the vicinity of aerosol surfaces, it is important to estimate both the three-dimensional (3-D) distributions of number concentrations and the coincidence of sulfate and mineral aerosols in order to represent it.

For absorbing aerosols, the direct radiative effect strongly depends on their altitude. Tegen and Lacis [1996] show that for a given optical thickness over a given surface, mineral aerosol can have a positive or negative radiative forcing depending on the altitude of the dust layer. It is therefore necessary to reproduce well both the horizontal and the vertical distribution of the aerosols. A discussion of the vertical distribution of mineral aerosol can be found in the work of Duce [1995]. This distribution consists over the oceans of structured layers that extend above the marine boundary layer up to altitudes of 7 km. This pattern of aerosol distribution is confirmed by lidar measurements [Iwasaka et al., 1983, 1988; Swap et al., 1992; Chazette et al., 1997; Dulac et al., 1997; Hamonou et al., 1997]. Vertical profiles of aerosols from biomass burning have been collected by differential absorption lidar (DIAL) during the NASA Global Tropospheric Experiments/Amazon Boundary Layer Experiment (GTE/ABLE 2A) [Browell et al., 1988] which took place over the Amazon Basin in July-August 1985. These vertical profiles showed considerable variations. The Transport and Atmospheric Chemistry near the Equator-Atlantic (TRACE A) experiment, which took place over the tropical South Atlantic in October 1992, showed enhanced aerosol loadings as a result of biomass burning. The UV-DIAL profile allowed to identify the vertical extent of the aerosol plumes over these regions [Anderson et al., 1996].

In a previous paper [Guelle et al., 1998] we presented a 1 year simulation of atmospheric $^{210}$Pb using a global aerosol transport model, and we extensively compared the model results to ground-based observations of concentrations and deposition. Lead 210 is a tracer of submicron size aerosol particles, which are principally removed from the atmosphere by wet scavenging [Balkanski et al., 1993]. In this paper we present a detailed comparison of model simulations using our scheme and two other wet-scavenging schemes from the literature in order to focus on the influence of wet-removal parameterization on aerosol distribution and deposition. Available observations are used as a reference, with special emphasis on vertical profiles.

Section 2 describes how the three scavenging schemes, used to compute aerosol global distributions, differ. Section 3 highlights the differences in deposition and $^{210}$Pb surface concentrations brought about by the different assumptions in scavenging. We then compare measured vertical profiles to those predicted by the model runs. Altitudinal measurements of $^{210}$Pb are used in sensitivity studies to discuss the assumptions made in the scheme presented by Guelle et al. [1998].

2. Description of the Atmospheric Model and of the Three Scavenging Schemes

2.1. Atmospheric Model

In this study we use the 3-D atmospheric model of tracer transport TM2 developed by M. Heimann at the Max-Planck-Institut für Meteorologie in Hamburg [Heimann and Keeling, 1989; Heimann, 1995]. It is an off-line model driven by the analyzed 12 hourly fields from the European Centre For Medium-Range Weather Forecasting (ECMWF). This version of the model has a $2.5^\circ \times 2.5^\circ$ horizontal resolution [Ramonet, 1994], nine sigma layers extending from the ground to 10 mbar, and is run with a 1 hour time step. Horizontal and vertical advection are computed using the slopes scheme of Russell and Lerner [1981], with a slope limitation added to avoid negative concentrations [Schulz et al., 1998]. Subgrid-scale vertical exchange processes are performed using the Louis [1979] formulation for turbulent vertical transport and a simplified version of the Tiedtke [1989] scheme to compute the mass fluxes in cumulus clouds.

The 3-D global simulations were conducted for the year 1991 and part of the year 1994 (January through March) when the $^{210}$Pb vertical profiles were measured. The simulation of the whole year 1991, which is used for comparison with ground-based measurements, is described by Guelle et al. [1998] for one of the wet-scavenging schemes and has been repeated with the other two. Every simulation presented in this work has been initialized with an empty atmosphere and started 2 months before the period of study. For the profiles sampled during September-October 1991 and February-March 1994, the runs were started on July 1, 1991, and December 1, 1993, respectively, and lasted 4 months. These simulations have been computed the same way as for the whole year 1991, with some slight differences. First, since we did not dispose of ECMWF ground precipitation fields for 1994, we have used the precipitation fields from the National Centers for Environmental Predictions/National Center for Atmospheric Research (NCEP/NCAR) reanalysis. While the 24 hour forecast precipitation fields from ECMWF for 1991 have a temporal resolution of 12 hours, the NCEP is 6 hourly. Both precipitation fields were regridded to the model resolution. Since we compare the model results to profiles measured over a few hours, the model outputs of these two shorter simulations were sampled every 6 hours.

Lead 210 is produced from $^{222}$Rn. In the simulations of fall 1991 and spring 1994, when we compare individual vertical profiles, we used the daily concentrations of $^{222}$Rn obtained from a previous simulation [Ramonet and Monfray, 1996] to compute the $^{210}$Pb source in the model. The same run was repeated with the three scavenging schemes described below. Details about the treatment of the size distribution can be found in the work of Schulz et al. [1998]. In the simulations pre-
sented, we have set the size distribution parameters of ambient aerosol carrying $^{210}$Pb to 0.4 μm for the mass median diameter and 1.9 for the geometric standard deviation [Sanak et al., 1981; Bondietti et al., 1988]. The details of the $^{222}$Rn and $^{210}$Pb simulations can be found in the work of Guelle et al. [1998], who compared extensively the $^{210}$Pb distribution obtained to available surface measurements of concentration and deposition. Here we focus on the description of the differences between the three different scavenging schemes used.

2.2. Wet-Scavenging Schemes

The first scheme has been developed by Kasibhatla et al. [1991] and has been adapted to the TM2 model by Rehfeld and Heimann [1995] to compute scavenging of radioactive tracers (hereinafter referred to as RH95). The second scheme, described by Walton et al. [1988] (hereinafter referred to as W88), has been used to scav-enge fire smoke [Ghan et al., 1988], nitrogen [Penner et al., 1991], carbonaceous aerosol [LIOUSSE et al., 1996], and $^{210}$Pb aerosol [Lee and Feichter, 1995]. Scheme RH95 was chosen for its simple treatment of wet scavenging. Scheme W88 is widely used in global aerosol simulations and has proven to give the best agreement with ground-based observations in a previous intercomparison paper of aerosol wet-scavenging schemes [Lee and Feichter, 1995]. Scheme 3 is the scheme used by Balkansky et al. [1993] for global simulation of $^{210}$Pb aerosol which has been expanded to deal with size-distributed aerosols [Guelle et al., 1998]. It has been applied to mineral aerosol studies [Guelle, 1998] and is described by Guelle et al. [1998] (hereinafter referred to as the B93-G98 scheme). Table 1 summarizes how the scavenging efficiencies are computed in these three different schemes.

2.2.1. Scavenging by synoptic precipitation. The three schemes compute scavenging by synoptic precipitation through first-order loss operators. In scheme RH95 the scavenging efficiency depends on the ground precipitation rate, which is equivalent to assume a constant scavenging ratio. Precipitating cloud heights are fixed to 3.6 km and do not allow the scavenging of the aerosol above. A buildup of the aerosol load occurs above 3.6 km altitude in regions where large-scale precipitation dominates. Schemes W88 and B93-G98 are similar in their formulation of synoptic scavenging. They are both based on the work of Giorgi and Chameides [1986], which accounts for the vertical distribution of precipitation. The vertical profiles used for synoptic precipitation are those from the GISS (Goddard Institute for Space Studies) general circulation model [see Guelle et al., 1998]. Only the B93-G98 scheme computes separately below-cloud scavenging. While scheme RH95 does not separate below- and in-cloud scavenging, scheme W88 does not compute it at all, which is of little impact for global modeling of small particles such as $^{210}$Pb carriers [Guelle et al., 1998], but could introduce a significant bias for large aerosols with diameters greater than 1 μm. In the case of partial evaporation the aerosol is not released but rather we assume that the raindrop diameter is reduced. Part of the raindrops might undergo complete evaporation and release their nucleated particle in the atmosphere, although physically a reduction in droplet diameter is more likely to occur. The sensitivity of the B93-G98 scheme to this assumption is discussed in section 3.

Table 1 shows that the formulations of the scavenging efficiency from schemes RH95 and W88 are similar since scavenging coefficients and the precipitating fractions are not much different. Therefore the only difference is the rain amount taken into consideration in the scavenging at any height z. Scheme W88 uses the local precipitation formation rate at height z, which represents only a fraction of the ground precipitation rate used by scheme RH95 at each altitude. Therefore these two scavenging schemes will lead to different aerosol vertical distributions, with more aerosol in scheme W88 below 3.6 km and more aerosol above 3.6 km in scheme RH95 since no scavenging occurs above that height.

Both scavenging schemes, W88 and B93-G98, take into consideration the vertical distribution of precipitation to scavenge aerosol. Scheme B93-G98 uses local precipitation formation rates to compute cloud water condensation rate, whereas W88 computes scavenging efficiency as a function of the formation of precipitation at height z. To compare schemes W88 and B93-G98, we computed the scavenging efficiencies of both schemes for different ground precipitation rates. The scavenging efficiencies computed by B93-G98 are at least twice that calculated for scheme W88 at any height. We conclude that scheme B93-G98 is more efficient at scavenging aerosol by synoptic precipitation than the other two schemes.

2.2.2. Scavenging by convective precipitation. Scheme B93-G98 computes aerosol scavenging in convective precipitation as part of the mass flux entrained in convective cloud. The other two schemes use a first-order loss coefficient with the same formulation as for the synoptic precipitation. Scheme RH95 uses a constant scavenging coefficient up to 8.6 km. In the scheme W88, scavenging depends on the rate of formation of precipitation, but no scavenging occurs below cloud base. The vertical profiles needed in scheme W88 for convective precipitation are the ones derived from the GISS model.

The main differences between the two schemes RH95 and W88 are as follows: in scheme W88, scavenging by convective precipitation is limited to 30% of the aerosol mass in the grid box since $F=0.3$ (see Table 1). For scheme RH95, all the aerosol mass in a given grid box can be scavenged, but the scavenging coefficient is 10 times less than for the W88 scheme. Supposing that
Table 1. Formulation of Scavenging Efficiency by Both Synoptic and Convective Precipitation for Three Schemes

<table>
<thead>
<tr>
<th>Scheme</th>
<th>Formulation</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>RH95</td>
<td>( \eta = 1 - \exp(-\Lambda P_s \Delta t) ) with ( \Lambda = 0.185 \text{ mm}^{-1} )</td>
<td>Scavenging is constant with altitude below 3.6 km. No scavenging above.</td>
</tr>
<tr>
<td>W88</td>
<td>( \eta = F \left[ 1 - \exp(-\beta \Delta t) \right] ) with ( F = 1 ) and ( \Lambda = 0.24 \text{ mm}^{-1} )</td>
<td>Scavenging is computed according to local formation of precipitation. Below-cloud scavenging is neglected.</td>
</tr>
<tr>
<td>B93-G98</td>
<td>If ( R &gt; 0 ) (in-cloud scavenging), ( \eta = F \left[ 1 - \exp(-\beta \Delta t) \right] ) with ( F, \beta ) function of ( Q ) after GC86. If ( R \leq 0 ) (below-cloud scavenging), ( \eta = F \left[ 1 - \exp(-\Delta \Delta t) \right] ) with ( F = f_{\text{cloud base}} ) and ( \Lambda ) depends on aerosol size distribution after DH76.</td>
<td>Below-cloud scavenging is computed according to local precipitation and aerosol size distribution.</td>
</tr>
</tbody>
</table>

Scavenging by Synoptic Precipitation

Scavenging by Convective Precipitation

Scavenging is constant with altitude below 8.6 km. No scavenging above.

Scavenging is computed according to local formation of precipitation. Below-cloud scavenging is neglected.

Scavenging is computed as part of the mass fluxes entrained in the cumulus updrafts according to convective precipitation occurrence.

The scavenging efficiency \( \eta \) is the removal rate of aerosol which corresponds to the ratio of the lost aerosol mass within a time step \( \Delta t \) (hour) to the initial mass. It depends on a scavenging coefficient \( \lambda (\text{mm}^{-1}) \) or \( \beta (\text{s}^{-1}) \) and, according to the wet-scavenging scheme used, on the precipitating fraction of the grid box \( F \), combined with either the ground precipitation rate \( P_s (\text{mm h}^{-1}) \), or the local precipitation rate \( P (\text{mm h}^{-1}) \), or the local precipitation formation rate \( R (\text{mm h}^{-1}) \), or the local condensation rate \( Q (\text{kg m}^{-2} \text{s}^{-1}) \), or the aerosol mass fraction entrained in the updrafts \( F_u \). References given in this table are Dana and Hales [1976], Giorgi and Chameides [1986], Tiedtke [1989], and Balkanski et al. [1993].

The local precipitation rate is one tenth of the ground precipitation, the two scavenging schemes will compute equal efficiencies up to 8 mm h\(^{-1}\). According to the GISS vertical profiles, most of the convective precipitation is forming between 1.4 and 10.3 km altitude, corresponding to four model layers in the TM2. Partitioning the ground precipitation rate among these four layers gives precipitation formation rates greater than the tenth of the ground precipitation rate in each of them, implying that scheme W88 is more efficient than scheme RH95, except below 1 km where it does not compute scavenging at all. Since mass fluxes (used by scheme B03-G98) and rainfall rates (used by the other two schemes) are not related in any simple way, we cannot simply discuss the effects of the respective formulations of these two different types of wet-scavenging schemes. Respective results will be compared hereinafter.

3. Results and Discussion

3.1. Deposition

Table 2 presents the annual mean deposition fluxes for \(^{210}\text{Pb} \) as calculated from the three wet-scavenging schemes. Both total deposition and deposition from synoptic and convective precipitation are presented by averaging over each 30° latitudinal band. The differences in surface deposition fluxes between the three schemes are small, ranging from about 20 Bq m\(^{-2}\) yr\(^{-1}\) at high northern latitudes to less than 10 Bq m\(^{-2}\) yr\(^{-1}\) in the tropics and southern hemisphere. Schemes RH95 and W88 agree closely, whereas the B93-G98 scheme leads to higher values at high and midlatitudes and to slightly lower values in the tropics.

In order to determine whether deposition measurements could allow to distinguish between these schemes, we have compared the annual total deposition fluxes simulated by the three schemes at the 147 sampling stations presented by Guelle et al. [1998]. Results are shown in Figure 1. We clearly see that the deposition fluxes predicted by schemes RH95 and W88 are almost equal and very similar to those simulated by scheme B93-G98. The only noticeable differences between the three schemes (highest values in Figure 1) occur at stations experiencing strong convective precipitation associated with the monsoon regime (India and Japan). These conclusions agree with Lee and Feichter [1995]. They found very similar yearly mean biases between \(^{210}\text{Pb} \) deposition fluxes observed and simulated by four
### Table 2. Zonal Averages of Annual $^{210}$Pb Deposition by 30° Wide Latitudinal Band for Each of the Three Schemes and for Each Deposition Type

<table>
<thead>
<tr>
<th>Latitude</th>
<th>Scheme RH95</th>
<th>Scheme W88</th>
<th>Scheme B93-G98</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Conv.</td>
<td>Syno.</td>
<td>Total</td>
</tr>
<tr>
<td>90°N-60°N</td>
<td>11</td>
<td>66</td>
<td>96</td>
</tr>
<tr>
<td>60°N-30°N</td>
<td>47</td>
<td>65</td>
<td>133</td>
</tr>
<tr>
<td>30°N-0°N</td>
<td>75</td>
<td>5</td>
<td>97</td>
</tr>
<tr>
<td>0°S-30°S</td>
<td>13</td>
<td>14</td>
<td>29</td>
</tr>
<tr>
<td>30°S-60°S</td>
<td>1</td>
<td>6</td>
<td>7</td>
</tr>
<tr>
<td>Global</td>
<td>42</td>
<td>21</td>
<td>75</td>
</tr>
</tbody>
</table>

Conv., convective; Syno., synoptic.

* Flux due to convective precipitation.

b Flux due to synoptic precipitation.

c The difference between the total deposition and the sum of the two different wet deposition fluxes gives the turbulent dry deposition contribution.

Figure 1. Comparison between the annual total deposition flux ($\text{Bq m}^{-2} \text{yr}^{-1}$) simulated by the three schemes at 122 stations, with differences ranging from 6.0 to 9.6%.

Figure 2 compares the residence time of $^{210}$Pb for each 30° latitudinal band and the $^{210}$Pb content of the free troposphere and of the boundary layer. We define the free tropospheric residence time as the ratio of the total free tropospheric content divided by the total deposition at the surface. In scheme W88 the residence times are the same for both the tropics and the mid-latitudes in both hemispheres. Looking at the tropics where convective precipitation dominates and at high latitudes where most precipitation is synoptic, it is clear that the most efficient scheme at scavenging in the tropics is W88, whereas B93-G98 is the most efficient at high latitudes. Table 2 and Figure 2 show that different partitionings of the three removal processes at midlatitudes can lead to the same residence times. For example, at northern midlatitudes, schemes RH95 and W88 both have nearly the same total deposition and residence times but with very different contributions from convective and synoptic precipitations. It is noteworthy that a negative feedback operated in the deposition: weaker scavenging will increase aerosol tropospheric concentration, which in turn increases the total deposition. Over long periods this feedback evens out differences in total deposition fluxes between the two schemes. Given all the above remarks and the small differences in total deposition seen in Figure 1, we conclude that a comparison between simulated and observed deposition alone is of limited use to assess the accuracy of a scavenging scheme.

### 3.2. Surface and Vertically Integrated Concentrations

Figure 3 displays the global distribution of the annual mean tropospheric $^{210}$Pb concentrations. In Table 3 we present surface and tropospheric annual mean concen-
We have compared the predicted monthly averaged concentrations in surface air from each of the three simulations with those observed at several stations. It is important to note that we only present results at the stations, among the 35 presented by Guelle et al. [1998], for which there are the most significant differences between the monthly concentrations simulated with the three schemes. Results are shown in Figures 4a-4c. We grouped the high latitudes together in Figures 4a (northern hemisphere) and 4b (southern hemisphere) to focus on the treatment of synoptic precipitation. The scheme W88 overestimates the observed concentrations by a factor of 2, whereas the other two schemes predict concentrations in much closer agreement with the observed values. If we consider annual concentrations in surface air at the 59 stations presented by Guelle et al. [1998] located poleward of 40° latitude, the mean bias against observed values is -4% with scheme B93-G98, but +10% with scheme RH95 and +32% with scheme W88 (bias(%)=100 x (concentrations simulated with the three schemes for each 30° wide latitudinal band (the troposphere is defined from the ground to 16, 10, and 9 km for latitudinal bands 0°-30°, 30°-60°, and 60°-90°, respectively). Different scavenging schemes lead to large differences in either tropospheric or surface concentrations compared to much smaller differences in surface deposition (see Table 2). At high latitudes the schemes RH95 and W88 predict concentrations greater by more than a factor of 2 compared to B93-G98. Moreover, whereas schemes B93-G98 and RH95 lead to similar surface concentrations at northern and southern midlatitudes, scheme W88 leads to larger concentrations. Close agreement among the three schemes is found in the tropics. We also note that the aerosol atmospheric loading in scheme RH95 is greater than the other two schemes for all latitudinal bands, due to higher upper tropospheric concentrations. This buildup yields to an elevated mean annual tropospheric residence time (9.5 days compared to 7.2 days estimated by Turekian et al. [1977] and 6.5 days by Lambert et al. [1982]). With the other two schemes (B93-G98 and W88), 210Pb has a residence time of 7.2 and 7.8 days.

Surface level concentrations (Table 3) differ mostly at high latitudes. Although the three schemes lead to similar surface concentrations in the tropics, scheme W88 yields higher concentrations than the other two deposition schemes at middle and high latitudes. In all three cases the vertical gradient of 210Pb shows increased concentrations with altitude between 60°S and 90°S because of the remote location of sources at these latitudes. It is noticeable that on a global average, tropospheric concentration is about 35% larger with scheme RH95 than with the other two schemes due to the limited height where scavenging takes effect. At high southern latitudes, scheme W88 predicts surface concentrations 20% higher than the other two schemes.
Table 3. Zonal Averages of Mean Annual $^{210}$Pb Concentration in Both Surface Air and Troposphere by 30° Wide Latitudinal Band for Each of the Three Schemes

<table>
<thead>
<tr>
<th>Latitude</th>
<th>Scheme</th>
<th>Scheme</th>
<th>Scheme</th>
<th>Scheme</th>
<th>Scheme</th>
<th>Scheme</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>RH95</td>
<td>W88</td>
<td>B93-G98</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>90°N-60°N</td>
<td>502</td>
<td>546</td>
<td>241</td>
<td></td>
<td>489</td>
<td>718</td>
</tr>
<tr>
<td>60°N-30°N</td>
<td>505</td>
<td>434</td>
<td>276</td>
<td>539</td>
<td>676</td>
<td>492</td>
</tr>
<tr>
<td>30°N-0°N</td>
<td>341</td>
<td>239</td>
<td>208</td>
<td>476</td>
<td>599</td>
<td>501</td>
</tr>
<tr>
<td>0°S-30°S</td>
<td>139</td>
<td>95</td>
<td>73</td>
<td>88</td>
<td>111</td>
<td>89</td>
</tr>
<tr>
<td>30°S-60°S</td>
<td>131</td>
<td>89</td>
<td>65</td>
<td>26</td>
<td>51</td>
<td>17</td>
</tr>
<tr>
<td>60°S-90°S</td>
<td>287</td>
<td>214</td>
<td>203</td>
<td>333</td>
<td>388</td>
<td>331</td>
</tr>
<tr>
<td>Global</td>
<td>280</td>
<td>214</td>
<td>203</td>
<td>489</td>
<td>718</td>
<td>390</td>
</tr>
</tbody>
</table>

The troposphere includes the atmosphere between the surface and the tropopause for which the altitude is defined according to Lambert et al. [1982] as 16, 10, and 9 km for latitudinal bands 0°-30°, 30°-60°, and 60°-90°, respectively.

The largest difference between schemes RH95 and B93-G98 appears at the Antarctic station Dumont d'Urville (67°S). It can be explained by the lack of scavenging by synoptic precipitation above 3.6 km in scheme RH95, hence the $^{210}$Pb in the free troposphere is scarcely scavenged and can easily reach high southern latitudes.

To compare the scavenging schemes in the tropics where convective precipitation is dominant, we considered six tropical stations in Figure 4c. Notwithstanding Midway and Mauna Loa stations, we see little differences in the concentrations simulated by the three schemes. Springtime concentrations at Midway are overpredicted by schemes RH95 and W88. This is the time of the year when Asian influences are most felt across the North Pacific [Merrill et al., 1989; Harris and Kahl, 1990], coincident with large-scale precipitation. The picture is reversed at Mauna Loa where these two schemes reproduce the spring maximum when the scheme B93-G98 shows no seasonal variation associated with this time of year. It is noteworthy that concentra-
tions at Mauna Loa (3400 m above sea level (asl)) during springtime are significantly higher than at Midway, although the distance from the Asian continent (where most of the $^{210}$Pb source is) to Mauna Loa is twice that of Midway. Clearly, our modeling does not allow to resolve these discrepancies. An underestimated source of $^{222}$Rn, the precursor of $^{210}$Pb, over Asia has been suggested to explain the differences at Mauna Loa (P. S. Kasibhatla and N. M. Mahowald, personal communication).

In summary, aside from the local differences depicted in this section and that the tendency scheme W88 has to overpredict high-latitude surface concentrations, the three schemes behave rather similarly at reproducing the monthly $^{210}$Pb concentrations measured near the surface at most of the sampling stations.

3.3. Vertical Distribution

Figure 5 depicts the respective zonal mean $^{210}$Pb concentrations as a function of altitude using the three scavenging schemes. The $^{210}$Pb at southern middle and high latitudes is transported from more northward regions where land covers much larger areas and hence where $^{210}$Pb is much more abundant. This explains the steep north-south gradient encountered in the southern hemisphere. At northern middle and high latitudes, since scheme W88 removes less efficiently the aerosol than the other two schemes, it produces higher surface concentrations. Another significant difference that can be seen is the unrealistically high zonal mean concentrations produced by scheme RH95 in the middle troposphere and above. Since scavenging occurs only up to 3.6 km height in that scheme, the $^{210}$Pb formed above contributes to these high values. In the tropics and subtropics the main difference is between the B93-G98 scheme which computes scavenging from the aerosol mass flux entrained in the updrafts, and the other two schemes that depend on the precipitation rates. Unlike the other two schemes the scheme B93-G98, based on mass fluxes, does not show a pronounced minimum concentration around 8 km height. Below, we further compare the model vertical profiles to observations in the tropics.

3.3.1. Comparison between observed and simulated $^{210}$Pb vertical profiles. To understand the influence of the different schemes on the vertical distribution, we have compared the profiles of concentrations issued from the three simulations with existing tropospheric measurements. An attempt to compare observed and simulated profiles with four different scavenging schemes is described by Lee and Feichter [1995].

![Figure 4c. Same as Figure 4a but for six stations located in the tropics.](image)

![Figure 5. Zonal averages as a function of altitude of mean annual $^{210}$Pb concentration ($\mu$Bq m$^{-2}$) obtained with schemes RH95 (top), W88 (middle), and B93-G98 (bottom). Isolines are 25, 50, 100, 150, 200, 300, 400, 500, and 600.](image)
Many of the measurements they used were taken in the stratosphere, and the comparison is complicated by the poor model resolution at these heights and the difficulty to resolve the stratosphere-troposphere exchange.

The shaded area in Figure 6 shows the location of observations from Moore et al. [1973] over North America. We have averaged these profiles from two periods: January and August. For each month there were only two instantaneous observed profiles, and in the absence of meteorological data for 1971, we used the 1991 simulation to compare mean January and August vertical profiles over the model grid box where the measurements took place. Vertical profile comparisons are presented in Figure 7. Only scheme B93-G98 reproduces the observed winter profile when synoptic precipitation dominates, although it overestimates the high tropospheric concentrations by approximately a factor of 2. No scheme reproduces accurately the slope of the observed summer profile. This could be due either to a different meteorology in 1971 than during the year 1991 of the simulation or to a poorly resolved convection in the model. Indeed, Mahowald et al. [1995] have shown, by comparison of predicted and observed vertical profiles of $^{222}$Rn concentration, that the Tiedtke [1989] scheme used in the TM2 causes too high convection during summer over North America.

To do a more in-depth study of the differences between the scavenging schemes, we compare the model results for the simulations of fall 1991 and spring 1994 to the vertical profiles taken by J. E. Dibb in Sept.-Oct. 1991 during the NASA Pacific Exploratory Mission-West, Phase A (PEM-West A) expedition [Dibb et al., 1996] and in Feb.-March 1994 during the PEM-West B expedition [Dibb et al., 1997], both taking place mostly over western North Pacific. In all cases we are able to compare these profiles to the model results for the analyzed meteorological conditions of the same days. The comparison to the observed vertical profiles of concentrations consists in selecting the locations included in the same model grid column where the plane flew at different heights during the same flight, i.e., approximately within 6 hours. The output concentrations in the model are set every 6 hours in order to compare as accurately as possible observed and simulated instantaneous concentrations. We show in Figure 6 the location where vertical profiles were measured (the plane ascents...
and descents do not allow to have profiles along all the tracks that were flown). We could assemble eight profiles for PEM-West A, and thirteen for PEM-West B. Figures 8a-8c show the comparison of the vertical profiles obtained by the three schemes with those observed; Figure 8a is for PEM-West A and Figures 8b and 8c for PEM-West B.

The PEM-West A campaign took place in late summer to early fall, when convective activity dominates over the Pacific. As already discussed, we can notice in Figure 8a that predicted concentrations with scheme B93-G98 are often higher than with the other two schemes, indicating a less efficient scavenging by convective precipitation. Leaving profile 2 for which the discrepancy between observed and predicted concentrations seems not to depend on the scavenging scheme, we see in Figure 8a that the scheme B93-G98 is in better agreement with observations than the other two schemes for all flights except for flight 7.

The comparison between two profiles (1 and 3) taken over the same location shows a threefold increase of the $^{210}$Pb concentrations at 8.5 km altitude. This is the result of a stratospheric air intrusion which penetrates deeply in the troposphere [see Browell et al., 1996, Plate 2]. The model does not account for this kind of problem because of its very poor vertical resolution in the stratosphere, preventing from simulating accurately the troposphere-stratosphere exchanges. This explains why this tropopause fold is not simulated by the model, yielding an underestimate of the high-altitude $^{210}$Pb concentration of profile 3.

The PEM-West B campaign gives an additional information since it took place at a time of the year (late winter to early spring) when synoptic precipitation is important at midlatitudes. Moreover, circulation patterns were also different than for the PEM-West A period, with the Japan jet responsible for most of the Asian continental outflow extending farther south and being stronger in intensity in Feb.-Mar. than in Sep.-Oct. [Gregory et al., 1997; Merrill et al., 1997]. Considering the convective and synoptic precipitation amounts for that period according to the analyzed precipitations from NCEP, we can distinguish profiles obtained in regions where convective precipitation dominates (profiles 11 to 15, Figure 8b) versus regions where synoptic precipitation is more important (profiles 16 to 23, Figure 8c). Aside from profile 15 for which no scheme succeeds in reproducing the observed concentrations, schemes RH95 and W88 overpredict the profiles by roughly a factor of 2, whereas scheme B93-G98 comes in closer agreement with the observations. In contrast to the results obtained with PEM-West A measurements, concentrations predicted with scheme B93-G98 over convective areas are less than those with the other two schemes. This is attributable to the synoptic precipitation band that spreads from Japan to southeastern China and removes efficiently $^{210}$Pb en route from the Asian continental source to more tropical regions where convective precipitation dominates.

This efficiency at scavenging is clearly observed in regions where convective precipitation dominates (Figure 8c). Schemes RH95 and W88 overestimate the observed concentrations, whereas scheme B93-G98 shows a much better agreement, except for profile 17. Again, the disagreement shown by scheme B93-G98 for profile 21 is attributable to a strong tropopause fold the model does not simulate. This conclusion was reached by Dibb et al. [1997] looking at O$_3$ and aerosol-scattering vertical profiles from a UV-DIAL instrument. These vertical distributions clearly show a deep intrusion of stratospheric air which can explain the twofold increase of observed $^{210}$Pb concentration at 10.7 km altitude between profiles 21 and 23 which were taken within 6 hours.

It is evident from these different profile comparisons that schemes RH95 and W88 are not efficient enough in removing $^{210}$Pb from the atmosphere via convective precipitation. However, we can notice in Figures 7 and 8c the reverse trend between middle/high and low troposphere for the profiles simulated by these two schemes. In fact, whereas scheme RH95 exhibits greater $^{210}$Pb concentrations in the high troposphere due to the lack of scavenging above 3.6 km altitude, it shows smaller $^{210}$Pb concentrations in the low troposphere, because of its more efficient scavenging since it does not distin-
Figure 8a. Comparison of instantaneous vertical profiles of $^{210}\text{Pb}$ concentrations ($\mu$Bq m$^{-3}$) predicted by the three schemes with vertical profiles observed over the western North Pacific, in September-October 1991 during the PEM-West A campaign. The different lines correspond to the cases in Figure 7. The observed profiles are plotted as solid circles for the measurements, as well as linear interpolation lines between two consecutive samples in altitude. This line is dotted when two consecutive samples are distant by more than 5 km in altitude.

Figure 8b. Same as Figure 8a but in February-March 1994 during the PEM-West B campaign in open ocean.

3.3.2. Quantitative comparison. In order to quantify the ability of each of the three schemes to simulate the vertical distribution of $^{210}\text{Pb}$ we have first compared the vertical gradients of concentrations measured and simulated. For this purpose we have calculated 47 gradients of observed $^{210}\text{Pb}$ concentrations as a function of height for each couple of two consecutive altitudes of samplings deduced from all the profiles presented.
in Figures 8a-8c. We have repeated these calculations for the three simulations, the $^{210}$Pb concentrations for each sampling altitude being interpolated linearly between the two surrounding model levels. The observed and simulated vertical gradients are then binned into 100 $\mu$Bq m$^{-3}$ km$^{-1}$ wide intervals for the comparison presented in Table 4. These results indicate that the mass flux scheme to compute aerosol scavenging by convective precipitation reproduces better the gradients than the two first-order schemes.

To estimate the extent to which the different wet-scavenging schemes could underpredict or overpredict the observed altitudinal $^{210}$Pb concentrations, we have compared model results to all the samples (including those used to assemble the 21 vertical profiles) carried out over the western part of the North Pacific Ocean during the two PEM-West campaigns. The location of these samples is indicated in Figure 9. Table 5 presents the mean bias between observed and simulated $^{210}$Pb concentrations for three altitude ranges. Schemes RH95 and W88 underestimate the observed concentrations in Sep.-Oct. 1991 and overestimate them in the whole troposphere in Feb.-Mar. 1994. This pattern is more pronounced for scheme W88. We suggest that it results from a combination of excessive aerosol scavenging by convective precipitation (PEM-West A results) and a too weak scavenging by synoptic precipitation (PEM-West B results). For scheme B93-G98 we observe a reverse trend with a closer agreement to observations (the

<table>
<thead>
<tr>
<th>Scheme</th>
<th>Within Range$^a$</th>
<th>Out of Range$^b$</th>
<th>Opposite Gradient$^c$</th>
</tr>
</thead>
<tbody>
<tr>
<td>RH95</td>
<td>49</td>
<td>19</td>
<td>32</td>
</tr>
<tr>
<td>W88</td>
<td>51</td>
<td>23</td>
<td>26</td>
</tr>
<tr>
<td>B93-G98</td>
<td>72</td>
<td>13</td>
<td>15</td>
</tr>
</tbody>
</table>

Values refer to the percentage from 47 observed gradients binned into the following intervals: $[-400;-300]$, $[-300;-200]$, $[-200;-100]$, $[-100;0]$, $[0;100]$, $[100;200]$ expressed in $\mu$Bq m$^{-3}$ km$^{-1}$.

$^a$Simulated gradient is in the same interval as the observed one.

$^b$Simulated gradient is out of the interval but has the same sign as the observed one.

$^c$Simulated gradient is opposite to the observed one.
Figure 9. Location of measurements carried out up to 10 km over the western North Pacific during the two PEM-West campaigns.

Table 5. Mean Bias Between $^{210}$Pb Concentrations Observed and Simulated With three Schemes During PEM-West Campaigns for Different Altitude Ranges

<table>
<thead>
<tr>
<th>Altitude Range, km</th>
<th>N</th>
<th>Scheme RH95</th>
<th>Scheme W88</th>
<th>Scheme B93-G98</th>
</tr>
</thead>
<tbody>
<tr>
<td>0-2</td>
<td>17</td>
<td>-30</td>
<td>-27</td>
<td>20</td>
</tr>
<tr>
<td>2-6</td>
<td>9</td>
<td>-22</td>
<td>-53</td>
<td>6</td>
</tr>
<tr>
<td>6-10</td>
<td>28</td>
<td>3</td>
<td>-46</td>
<td>24</td>
</tr>
</tbody>
</table>

PEM-West A Campaign (Sept.-Oct. 1991)

PEM-West B Campaign (Feb.-March 1994)

N is the number of samples.

3.4. Sensitivity of Vertical Distribution to Assumptions of Scheme B93-G98

Several assumptions have been made in the scheme B93-G98 to compute the wet scavenging of aerosols and have been discussed by Guelle et al. [1998] through sensitivity runs. Two of these assumptions have to do with the treatment of scavenging by synoptic precipitation. Scheme B93-G98 considers both no interstitial aerosol within the cloud and that partial evaporation below the cloud does not allow any release of aerosol particles. Guelle et al. [1998] showed sensitivity simulations to estimate the validity of these assumptions. One simulation has been done with a 70% in-cloud scavenging efficiency (hereinafter referred to as simulation INC70) instead of 100% in the reference simulation, and the other one considers that a fraction of the aerosol in precipitation is released below the cloud, this fraction depending on the rainwater evaporation rate (hereinafter referred to as simulation EVAP). The previous study has shown that ground measurements were not sufficient to draw any conclusion, either because of the weak change in the surface concentration or deposition induced by the sensitivity runs or because of the lack of measurements in the areas where there is a significant difference between the reference and the sensitivity simulations. We looked here at whether sensitivity simulations can induce significant modification of the vertical distribution of aerosols which could be evaluated through the observed vertical profiles.

We have repeated these two sensitivity simulations, for the two periods of PEM-West campaigns with a 2 month spin up, and compared the predicted vertical profiles with those from both the reference simulation and the observations. We found in simulation INC70 an increase of the $^{210}$Pb ranging from 10 to 20% above 7 km altitude in Sept.-Oct. 1991 (PEM-West A), compared to the standard run, and a 5 to 35% increase in the whole tropospheric column in Feb.-March 1994 (PEM-West B), except for profile 17 which exhibits a twofold increase of the tropospheric $^{210}$Pb concentrations. For the simulation EVAP the increase in concentration ranges from 0 to 60% from the ground to 10 km altitude for the two campaigns.

Figure 10 shows the observed profiles and those from the reference and sensitivity simulations for the cases when the sensitivity simulations lead to a significant change in the vertical profile. For profiles 2, 6, and 17, no conclusion can be drawn from the comparison of reference and EVAP simulations with the observations. On the other hand, the profiles 5 and 18 show that the results from the reference simulation agree well with measurements near the surface, whereas simulation EVAP (which differs by its treatment of evapora-
Figure 10. Comparison of instantaneous vertical profiles of observed $^{210}$Pb concentrations ($\mu$Bq m$^{-3}$) with those simulated by the reference and EVAP simulations (see text) for the PEM-West A period (top) and those simulated by the reference EVAP and INC70 (see text) simulations for the PEM-West B period (bottom). Thick solid line corresponds to observations, thin solid line with solid squares to the reference simulation, dotted line with open squares to the EVAP simulation, and dashed line with open circles to the INC70 simulation. Only profiles for which the difference between reference and one of the sensitivity simulations is visible are plotted. The results of INC70 simulation are not plotted for PEM-West A period because of no significant change compared with the reference simulation.

Table 6. Same as Table 5 but for Sensitivity Simulations With Scheme B93-G98

<table>
<thead>
<tr>
<th>Altitude Range, km</th>
<th>N</th>
<th>Mean Bias, %</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Reference</td>
</tr>
<tr>
<td>0-2</td>
<td>17</td>
<td>20</td>
</tr>
<tr>
<td>2-6</td>
<td>6</td>
<td>6</td>
</tr>
<tr>
<td>6-10</td>
<td>28</td>
<td>24</td>
</tr>
<tr>
<td><strong>PEM-West B Campaign (Feb.-March 1994)</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0-2</td>
<td>23</td>
<td>-17</td>
</tr>
<tr>
<td>2-6</td>
<td>30</td>
<td>-6</td>
</tr>
<tr>
<td>6-10</td>
<td>43</td>
<td>-9</td>
</tr>
</tbody>
</table>

Evaporation (EVAP) overestimates them. This substantiates the preliminary results from Guelle et al. [1998] who found that computing a partial release of the aerosol through rainwater evaporation tends to overestimate the observed surface concentrations.

The profile 16 shows that the simulation INC70 simulates better than the reference simulation $^{210}$Pb concentrations at 6 and 9 km altitude. Unfortunately, only one profile improves the agreement with observations. We therefore also looked at the individual measurements depicted in Figure 9. We computed in Table 6 the mean bias between the observations and the model results for the reference and sensitivity simulations. We see the weak change in the middle and high troposphere when the treatment of evaporation is changed. When incorporation of aerosols into liquid water is reduced to 70% (INC70), the mean bias at reproducing PEM-West A
concentrations slightly increases, but there is a significant improvement at reproducing PEM-West B concentrations, with a mean bias equal to zero for the whole free troposphere.

4. Conclusions

A wet-scavenging scheme [Balkanski et al., 1993; Guelle et al., 1998] has been incorporated in a global 3-D atmospheric transport model which fully describes the aerosol size distribution [Schulz et al., 1998]. This scheme treats differently removal by convective from the removal by synoptic precipitation. While the former considers the aerosol mass entrained in wet convective updrafts, the latter is based on the vertical distribution of precipitation and computes separately in-cloud and below-cloud scavenging. Since wet removal below the cloud depends on particle size, we have chosen to use a parameterization that computes the aerosol size distribution before and after scavenging. The influence on the size distribution is particularly important for aerosol sizes above 1 μm diameter.

In order to assess improvements in aerosol wet deposition brought by our parameterization we have compared it with two other schemes widely used in global aerosol simulations: one in which the scavenging efficiency is uniform in the precipitating cloud from the top to the ground regardless of where precipitation is formed [Rehfold and Heimann, 1995], and the other which accounts for the variations with altitude of the precipitation formation but does not consider below-cloud scavenging [Walton et al., 1988]. Both of these schemes use first-order loss operators with different parameters for convective and synoptic precipitation.

We have compared the model results of simulations of 210Pb aerosol using these three schemes with observed concentrations in surface air and deposition. This has revealed that all the schemes allow us to reproduce well the observed fluxes of 210Pb annual deposition. One of the striking features of this study is obtained with the scheme RH95 which relies on simple assumptions. This scheme considers a constant scavenging efficiency throughout the precipitating column and allows the scavenging only below a constant altitude. In addition, it does not differentiate below-cloud from in-cloud scavenging. Nonetheless, we see no detectable advantage to use a more detailed scheme such as B93-G98 rather than this scheme since it reproduces both surface concentration and deposition.

To point out where the distributions of aerosols were different, we investigated the ability of the three schemes at reproducing 210Pb concentrations measured as a function of altitude. We have shown that only scheme B93-G98 is in agreement with most of the available observed vertical profiles of concentrations. We have compared the zonally averaged vertical distribution of aerosol obtained with the three schemes and found large discrepancies, emphasizing the need for more measurements of aerosol concentration in altitude (vertical profiles or airborne campaigns) to infer the accuracy of wet-scavenging schemes at the global scale. A strong conclusion from this work is that ground measurements taken alone are not sufficient to evaluate an aerosol wet-scavenging scheme.

Future simulation with mineral dust, for which the below-cloud scavenging coefficients are significantly higher, will permit to infer the importance of an improved parameterization of below-cloud scavenging for larger aerosol and to estimate the validity of our model to scavenge aerosols of larger size. Such a study is needed since the present work has shown the importance wet scavenging has on the vertical distribution of aerosols. The inability of representing vertical distribution will propagate into large uncertainties at estimating the radiative impact of aerosols, their lifetime, and transport to remote areas.

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References


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