Estimation of Fe and Al aerosol solubility from a diffusive method based on particle size

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Abstract: Mineral aerosol deposition is recognized as the dominant source of iron to the open ocean. Solubility of this aerosol iron is highly uncertain. Previous solubility estimates range widely from 0.01-80%. Here we use the diffusion equation to solve for the solubility of different particle sizes. We derived a time and spatial varying solubility map for aerosol Fe and Al based on the aerosol lifetime in the atmosphere and aerosol deposition for different size bins from a global dust model (DEAD). Our solubility map corresponds well with the observations from the CLIVAR/CO2 cruise in the North Atlantic Ocean. But it does not explain the large solubility variation observed from the 2002 IOC cruise in the North Pacific Ocean. We examine the impacts of spatially variable, size-dependent solubility on marine biogeochemistry with the Biogeochemical Elemental Cycling (BEC) ocean model. Simulated values of dissolved Al and Fe were compared with observations under constant and variable aerosol solubility. Diffusion-based variable solubility considerably reduces the global RMS error of the simulations.

Conceptual Model I: Diffusion in Solid Particles

Once the dust particles are mobilized into the atmosphere, condensation of water vapor takes place on the particle surface with increasing relative humidity. Based on experience relationship of relative humidity $f$ and equivalent radius $r^*$ (Hanel, 1976), $r^* = 1.335r$, at 80% relative humidity. The dust particle dissolution process begins at the condensation (Figure 1):

Step 1: aqueous dissociation reaction and diffusion of species A from the interface

Step 2: transfer of the species across the interface

Step 3: diffusion of species A inside the solid particle from center to surface

Step 3 decides the characteristic time because step3 is much slower than the first two steps.

Assume the particle is homogeneous and the particle radius is not changing during the dissolution process, we can describe step 3 using the following:

$$\frac{\partial N_A}{\partial t} = D_o \frac{\partial^2 N_A}{\partial r^2} + \frac{1}{r^3} \left( -N_A \right)$$

Here $N_A(r, t)$ is the concentration of species A; $r$ is the radius of the particle; $D_o$ is the initial concentration; $N_A(r, 0) = N_A(0, r)$; $D_o$ is the diffusion coefficient; and $N_A(r, t)$ denote concentration at particle surface.

Solving by standard separation of variables, apply to Fick’s Law again and integrate, we get the maximum fractional solubility of species A at time $t$ with:

$$\alpha = \frac{N_A(t, r)}{N_A(0, r)} = \sum \frac{1}{n^3} \left( -n^3 \frac{D_o}{r^3} \right)$$

Conceptual Model II: pH effect

The particles still take up water vapor and begin to dissolve as in the first model. The difference is we let the particle radius change with the dissolution in a function of acid concentration and time (Figure 2):

$$r(t) = r + \alpha \cdot \text{[acid]} \cdot t$$

Then the fractional solubility is:

$$S(t) = 1 - \frac{r^*(t) - r}{r^*}$$

Comparing the predicted solubility with observations, we found that it corresponds well with the observations from the CLIVAR/CO2 cruise and other data in the North Atlantic Ocean. But it does not explain the large solubility variation observed from the 2002 IOC cruise and others in the North Pacific Ocean (Figure 6).

Preliminary Results from Conceptual Model I: The diffusion coefficient varies in a large range from $10^{-12}$ cm$^2$/s to $10^{-40}$ cm$^2$/s. Compare to the observed dust dissolution time scale, we choose to use $10^{-12}$ cm$^2$/s in our model (Figure 3).

Discussion: Several factors that may be important but not included in Modellaire aerosol life time in the atmosphere, pH effect and particle origins. We plan to study them in the future. The solubility data were collected using different methods and whether the scavenging process is included is not clear.

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References: