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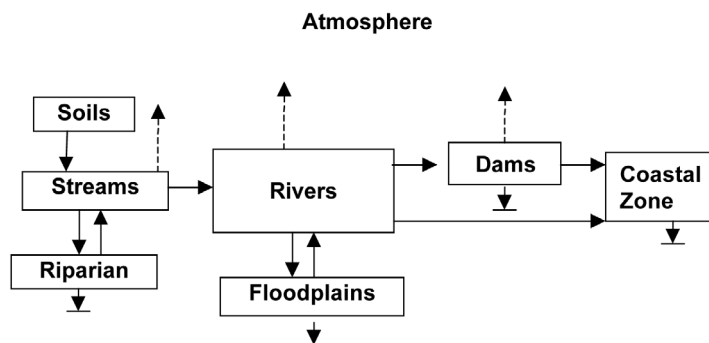
## Pathways of Atmospheric CO<sub>2</sub> through Fluvial Systems

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### Uncertainties in the Current “Global Fluvial Systems Model”

As the main pathway for the ultimate preservation of terrigenous production in modern environments, the transfer of organic matter from the land to the oceans via fluvial systems is a key link in the global carbon cycle (Ittekkot and Haake 1990; Degens et al. 1991; Hedges et al. 1992). Hence, the “role” of rivers in the global carbon cycle is most typically expressed as the fluvial export of total organic and dissolved inorganic carbon from land to the ocean (e.g., Likens et al. 1981). As will be discussed in more detail later in this chapter, the most common literature estimations of the magnitude of these fluxes are 0.4 petagrams of carbon per year (PgC y<sup>-1</sup>) for total organic carbon (evenly divided between particulate and dissolved organic phases), and 0.4 PgC y<sup>-1</sup> for dissolved inorganic carbon. While these bulk fluxes are small components of the global C cycle, they are significant compared to the net oceanic uptake of anthropogenic CO<sub>2</sub> (Sarmiento and Sundquist 1992) and to the interhemispheric transport of carbon in the oceans (Aumont et al. 2001).

This chapter examines and expands on several inconsistencies in this conventional model. The first is in the estimates of the flux quantities and in the relative influence of natural and anthropogenic processes in determining these fluxes. While much river research has emphasized concentrations of carbon, sediments, and/or nutrients, with a focus on export to the oceans (Meybeck 1982, 1991; Degens et al. 1991; Milliman and Syvitski 1992), considerable uncertainties remain. The second problem is that the role of fluvial systems may not be limited to fluvial exports to the coastal zone. Continental sedimentation may sequester large amounts of carbon in lower depressions and wetlands (Stallard 1998; Smith et al. 2001). More recent estimates indicate that CO<sub>2</sub> outgassing to the atmosphere from river systems may be an important pathway (Cole and Caraco 2001; Richey et al. 2002).



**Figure 17.1.** Major reservoirs and pathways of atmospheric  $\text{CO}_2$  in fluvial systems. Atmospheric  $\text{CO}_2$  fixed into streams and their riparian (near-stream) zones (as dissolved  $\text{CO}_2$  or organic matter) is stored as alluvium, released back to the atmosphere, or transported down stream networks to larger river systems. This now-riverine carbon exchanges with floodplains, is outgassed, or is retained behind dams. Finally the “remaining” fluvial carbon is exported to the coastal zone.

## Dynamics of Fluvial Systems

Fluvial systems integrate hydrological and biogeochemical cycles, over scales from small streams to regional and ultimately to continental basins. The key issue here is the quantity of carbon removed from the atmosphere relative to the amount returned to the atmosphere, along the overall fluvial pathways. This sequence of processes can be summarized in the form of a box model (Figure 17.1). Briefly, three primary forms of carbon of atmospheric origin are transported through fluvial systems. Particulate organic carbon (POC) enters rivers from the erosion of soils (typically older materials) and as leaf litter (typically newly produced). Dissolved organic carbon (DOC) is produced through solubilization of soil organic carbon and enters streams via groundwater. Total dissolved inorganic carbon (DIC) is produced via weathering, as the dissolution of carbonate and silicate rocks. This process sequesters atmospheric  $\text{CO}_2$ , establishes the alkalinity, and influences the pH of water, which governs the subsequent partitioning of DIC between  $\text{pCO}_2$ , bicarbonate, and carbonate ions. The dynamics of carbon in fluvial systems are not defined solely by the export fluxes of bulk C. Rather, they are defined as a complex interplay of multiple C fractions; each exhibits distinct dynamics and compositional traits that hold over very broad ranges of geological, hydrological, and climatic conditions (Hedges et al. 1994).

Evaluating the model of Figure 17.1 is a challenge. The dynamics are complex, and multiple time constants are involved. Data are scarce, particularly in many of the most anthropogenically affected systems. The distribution of the constituent processes varies dramatically across the face of the globe (with some of the most important regions being the least measured).

### *Mobilization from Land to Water and Riparian Zones*

The fluxes from land to rivers are generally inferred directly from the fluxes out of a basin, especially at a global scale. Although there is considerable truth to this for dissolved species (especially conservative ones), it is less true for particulate species, especially with human intervention.

The modern terrestrial sediment cycle is not in equilibrium (Stallard 1998). Meade et al. (1990) estimated that agricultural land use typically accelerates erosion 10- to 100-fold, via both fluvial and Aeolian processes. Multiple other reports in the literature support this conclusion. With the maturation of farmlands worldwide, and with the development of better soil conservation practices, it is probable that the human-induced erosion is less than it was several decades ago. Overall, however, there has been a significant anthropogenic increase in the mobilization of sediments (and associated POC) through fluvial processes. The global estimates of the quantities, however, vary dramatically. Stallard (1998) poses a range of scenarios, from 24 to 64 Pg y<sup>-1</sup> of bulk sediments (from 0.4 to 1.2 Pg y<sup>-1</sup> of POC). Smith et al. (2001) estimate that as much as 200 Pg y<sup>-1</sup> of sediment is moving, resulting in about 1.4 Pg y<sup>-1</sup> (using a lower percentage C than Stallard 1998).

Where does this material go? Does it all go downstream via big rivers, ultimately to the ocean, or is it stored inland? Stallard (1998) argues that between 0 and 40 Pg y<sup>-1</sup> of sediments (0 to 0.8 Pg C y<sup>-1</sup> POC) is stored as colluvium and alluvium and never makes it downstream. Smith et al. (2001), using a different approach, estimate that about 1 Pg C y<sup>-1</sup> of POC is stored this way. If this movement is merely transferring POC from one reservoir to another, with the same residence times, there is no net change in the C cycle. Then the issue is, to what degree can the remaining soils sequester carbon by sorption to the newly exposed mineral soils? Both Stallard (1998) and Smith et al. (2001) argue that carbon is removed from the upper portion of the soil horizon, where turnover times are relatively rapid (decades or shorter) into either of two classes of environments with longer turnover times: wetlands and smaller, deeper depositional zones, coupled with new carbon accumulation at either erosional or depositional sites. Both assume that oxidation of organic C in transit is minimal, and both use quite conservative values for total suspended sediment (TSS) export. If true, this sequence of processes would result in a significant C sink, on the order of 1 PgC y<sup>-1</sup>.

### *Within-River Transport and Reaction Processes*

Within-river transport processes carry these eroded materials downstream through the river network. Transport is not passive; significant transformations occur along the way. Rivers exchange with their floodplains (depending on how canalized and diked a river is). The movement of POC is, of course, directly linked to the movement of suspended sediments. Sediments are deposited and remobilized multiple times and over long timescales. In the Amazon, for example, Dunne et al. (1998) computed that as

much sediment was being recycled within a reach as was leaving it. Presumably, a significant amount of the erosion-excess sediment discussed in the previous section makes it some distance downstream but is then slowed and retained within the alluvial floodplains.

An additional process within flowing water significantly affects organic matter (OM)—the mineralization to  $p\text{CO}_2$ . Most river and floodplain environments maintain  $p\text{CO}_2$  levels that are supersaturated with respect to the atmosphere. High partial pressures of  $\text{CO}_2$  translate to large gas evasion fluxes from water to atmosphere. Early measurements in the Amazon suggested that global  $\text{CO}_2$  efflux (fluvial export plus respiration) from the world's rivers could be on the order of  $1.0 \text{ PgC y}^{-1}$ . Recent measurements of temperate rivers lead to estimates of global river-to-atmosphere (outgassing) fluxes of  $\sim 0.3 \text{ PgC y}^{-1}$ , which is nearly equivalent to riverine total organic carbon (TOC) or dissolved inorganic carbon (DIC) export (Cole and Caraco 2001). Richey et al (2002) computed that outgassing from the Amazon alone was about  $0.5 \text{ PgC y}^{-1}$ . Assuming that the fluxes computed for the Amazon are representative of fluvial environments of lowland humid tropical forests in general, surface water  $\text{CO}_2$  evasion in the tropics would be on the order of roughly  $0.9 \text{ PgC y}^{-1}$  (three times larger than previous estimates of global evasion). Factoring in the recent Amazon results, a global flux of at least  $1 \text{ PgC y}^{-1}$  directly from river systems to the atmosphere is likely.

What is the source of the organic matter being respired? Is it labile contemporary organic matter, recently fixed in the water by plankton or nearshore vegetation, or is it some fraction of the allochthonous (terrestrial) matter in transport? The prevailing wisdom is that riverborne organic matter is already very refractory and not subject to oxidation (after centuries on land). The “age” of riverine organic matter yields some important insights.

Measurements of the  $^{14}\text{C}$  ages of organic matter and  $\text{CO}_2$  in river water are very few, but the results are intriguing (Hedges et al 1986). Cole and Caraco (2001) found that the POC (and to a lesser degree dissolved organic carbon [DOC]) entering the Hudson is greatly depleted in  $^{14}\text{C}$ , suggesting that the particulate material was originally formed on average  $\sim 5,000$  years ago. Their analysis of  $\Delta^{13}\text{C}$  suggests that this material is of terrestrial origin and unlikely to be ancient marine sedimentary rocks. Furthermore, they found that organic matter (OM) pools became selectively enriched in  $^{14}\text{C}$  downstream. Based on an inverse modeling approach, they hypothesized that this enrichment is due to utilization of old organic carbon, with dilution by recent primary production. That is, organic C that had resided in soils for centuries to millennia, without decomposing, is then decomposed in a matter of a few weeks in the riverine environment (how this happens is an intriguing question in its own right). Their results are not unique to the Hudson. Raymond and Bauer (2001) found that four rivers draining into the Atlantic are sources of old ( $^{14}\text{C}$ -depleted) and young ( $^{14}\text{C}$ -enriched) terrestrial dissolved organic carbon and of predominantly old terrestrial particulate organic carbon. Much of the younger (relatively speaking) DOC can be selectively degraded over the res-

idence times of river and coastal waters, leaving an even older and more refractory component for oceanic export.

Thus, pre-aging and degradation may alter significantly the structure, distribution, and quantity of terrestrial organic matter before its delivery to the oceans. As noted by Ludwig (2001), the OM that runs from rivers into the sea is not necessarily identical to the OM upstream in river catchments. Cole and Caraco (2001) observe that the apparent high rate of decomposition of terrestrial organic matter in rivers may resolve the enigma of why OM that leaves the land does not accumulate in the ocean (*sensu* Hedges et al. 1997). Overall, this sequence of processes suggests that the OM that is being respired is translocated in space and time from its points of origin, such that, over long times and large spatial scales, the modern aquatic environment may be connected with terrestrial conditions of another time.

### *Input to Reservoirs*

Just because dissolved and particulate materials enter a river does not mean that they reach the ocean; modern reservoirs have had a tremendous impact on the hydrologic cycle. Starting about 50 years ago, large dams were seen as a solution to water resource issues, including flood control, hydroelectric power generation, and irrigation. Now, there are more than 40,000 large dams worldwide (World Commission on Dams 2000). This has resulted in a substantial distortion of freshwater runoff from the continents, raising the “age” of discharge through channels from a mean between 16–26 days and nearly 60 days (Vörösmarty et al. 1997). Whereas erosion has clearly increased the mobilization of sediment off the land, the proliferation of dams has acted to retain those sediments. Vorosmarty et al. (2003) estimates that the aggregate impact of all registered impoundments is on the order of 4–5 Pg y<sup>-1</sup> of suspended sediments (of the 15–20 Pg y<sup>-1</sup> total that he references). Stallard (1998) extrapolates from a more detailed analysis of the coterminous United States to an estimate of about 10 Pg y<sup>-1</sup> worldwide (versus 13 Pg y<sup>-1</sup> efflux to the oceans), for a storage of about 0.2 PgC y<sup>-1</sup> (which he includes as part of his overall calculation of continental sedimentation).

### *Export to the Coastal Zone*

The conventional wisdom is that the flux of POC and DOC are each about 0.2 PgC y<sup>-1</sup>, and DIC is 0.4 PgC y<sup>-1</sup> (e.g., Schlesinger and Melack 1981; Degens 1982; Meybeck 1982, 1991; Ittekkot 1988; Ittekkot and Laane 1991; Ludwig et al. 1996; Ver at al. 1999). That these analyses converge is not terribly surprising. They are all based on much of the same (very sparse) field data and use variations of the same statistically based interpolation schemes. Let us evaluate these numbers.

Because direct measurements are few, POC flux estimations are typically a product of the flux of total suspended sediments (TSS) and the estimated weight-percent

organic C (w%C) associated with the sediment (because the bulk of POC is organic C sorbed to mineral grains). The first problem is an adequate resolution of the TSS flux. Data on TSS are frequently poor and of unknown quality. Many reported data are surface samples, and the depth integrations necessary to accurately characterize sediment flux are on the order of two to three times higher. Additionally, much sediment moves during episodic storm events, when measurements are almost never made.

As summarized by Vörösmarty et al. (2003), estimates of TSS transport to the oceans have ranged from 9 Pg y<sup>-1</sup> to more than 58 Pg y<sup>-1</sup>, with more recent studies converging around 15 to 20 Pg y<sup>-1</sup>. These estimates are generally based on extrapolations of existing data, which are weighted to the large rivers of passive margins and temperate regions. Milliman and Syvitski (1992) called attention to the much higher yield rates from steep mountainous environments (without directly computing a global total). More recently, Milliman et al. (1999) estimated that the total sediment flux from the East Indies alone (the islands of Borneo, Java, New Guinea, Sulawesi, Sumatra, and Timor), representing about 2 percent of the global land mass, is about 4 Pg y<sup>-1</sup>, or 20–25 percent of the current global values. This type of environment (steep relief, draining directly to the oceans) is found elsewhere in the world, so the results are not likely to be unique. New data from Taiwan support these high levels, with isotopic analyses of the C showing that a significant part of the flux is human-driven (Kao and Liu 2002).

To obtain POC flux estimates, these values (and their uncertainties) must be multiplied by w%C. Meybeck (1991) divided particulate carbon into inorganic (PIC) and organic (POC) phases and assumed that high-sediment rivers have very low carbon fractions (0.5 w%C), representative of shale; he essentially does not consider the latter to be “atmospherically derived” and hence discounts it from estimates of fluxes to the ocean. More recent values for w%C tend to be in the 1–2 percent range, and higher for organic-rich systems (Richey et al. 1990; Stallard et al. [Delete et al.?] 1998; Gao et al. 2002).

To account for this range, POC flux can be computed as an ensemble based on different combinations of w%C and TSS fluxes, resulting in a range of 0.3 PgC y<sup>-1</sup> to 0.8 PgC y<sup>-1</sup>, with a “more likely” level of about 0.5 PgC y<sup>-1</sup> (depending on assumptions used). Therefore, it is possible that the common estimate of 0.2 PgC y<sup>-1</sup> is low and that the overall value lies in the range of 0.2–0.5 PgC y<sup>-1</sup>. The common estimate for PIC of 0.2 PgC y<sup>-1</sup> (Meybeck 1991; Ver et al. 1999) may also be underestimated, if sediment fluxes are higher.

The value of 0.2 PgC y<sup>-1</sup> for DOC export may also be low. DOC is also subject to sparse (and questionable) measurements, without the availability of a proxy like TSS for POC. Aitkenhead and McDowell (2000) developed a model of riverine DOC flux as a function of soil C:N. Using this model, they computed a global flux of 0.4 PgC y<sup>-1</sup>, or twice the common estimate. That is, the total organic C output from fluvial systems may well be approximately double the original estimates, in the ~0.8 PgC y<sup>-1</sup> range.

### *Marine Fate*

Long-term preservation of terrestrially derived organic matter in the oceans occurs largely within sediments that accumulate along continental margins. Organic carbon within these sediments is thought to be preserved largely because it is adsorbed to mineral grains (Keil et al. 1994; Mayer 1994; Bishop et al. 1992). Hedges and Keil (1995) estimated that carbon preservation along continental margins over the Holocene was split roughly evenly between sediments accumulating within the delta or sedimentary plume of rivers and non-deltaic sediments accumulating outside the direct influence of major rivers (but within range of multiple smaller systems).

The efficiency of storage between deltaic and non-deltaic systems is different. The amount of organic carbon in non-deltaic continental shelf sediments falls in a narrow range (0.5–1.1 milligrams of carbon per square meter [ $\text{mg C m}^{-2}$ ] of mineral surface), and typically >90 percent of the preserved organic matter is adsorbed to mineral surfaces. Deltaic sediments are distinctly different, containing only a fraction of the organic carbon (by weight) found in other margin sediments. Suspended sediments from the Amazon River, for example, have loadings ( $\sim 0.67 \text{ mg C m}^{-2}$ ) that are three times higher than the corresponding deltaic sediments (Keil et al. 1997), so more than two-thirds of the terrestrial particulate organic load delivered to the Amazon delta is lost from the mineral matrix and is not preserved. The Mississippi, Yellow, and other river/delta systems also show extensive loss of terrestrial organic matter. Thus many deltaic systems bury only a small fraction of the potential organic load normally sorbed to mineral particles, with the balance presumably desorbed or mineralized (and entering the DIC pool).

The organic matter lost by mineralization and not buried is one of the factors in maintaining the historical perspective that marginal seas are net heterotrophic (Chen, Chapter 18, this volume). But Chen (2003) reviews more recent evidence, based on direct measurements of  $\text{pCO}_2$  (again, showing the critical importance of actual field measurements of key parameters!) and comes to the conclusion that these seas are net autotrophic, driven primarily by nutrients delivered via upwelling (with enhanced nutrients delivered by rivers leading to eutrophication constituting only a minor source), and net consumers of atmospheric CO<sub>2</sub>. The overall implication of this sequence of processes is that much of the anthropogenically mobilized riverborne OM (and perhaps the naturally mobilized OM) is liable to remain in the marine environment over timescales longer than the current increase of atmospheric CO<sub>2</sub>.

### *Anthropogenic Transient*

To what degree have these fluxes been influenced or impacted by human activities—that is, how much of this carbon is an anthropogenic transient? The consensus is that human-induced erosion has dramatically accelerated the movement of sediments (and

POC). While some of this material “hangs up” on land (sedimentation, reservoirs), some of it likely escapes to the sea (some of the regions with highest sediment yields have very few dams). There is evidence that anthropogenic processes have an affect on DIC. Raymond and Cole (2003) report an increase in the alkalinity of the Mississippi, which implies an increase in the consumption of atmospheric  $\text{CO}_2$  through weathering. Jones et al. (2003), however, report a systematic decrease in  $\text{pCO}_2$  in rivers across the United States, which they attribute to large-scale declines in terrestrial  $\text{CO}_2$  production and import into aquatic ecosystems and not to terrestrial weathering or in-stream processes.

In the case of DOC, there is simply not enough information available to draw conclusions. Clair et al. (1999) suggested that DOC export from basins in Canada might increase by 14 percent with a doubling in atmospheric  $\text{CO}_2$ . An additional factor rarely addressed in rivers is direct loading from urban and industrial sources (Ver et al 1999; Abril et al. 2002). In evaluating the consequences of continental sedimentation and the potentially higher fluxes of POC, a net transient exported from land of roughly  $1 \text{ PgC y}^{-1}$  is possible, with perhaps half of that going to the sea and the other half divided equally between outgassing and sedimentation.

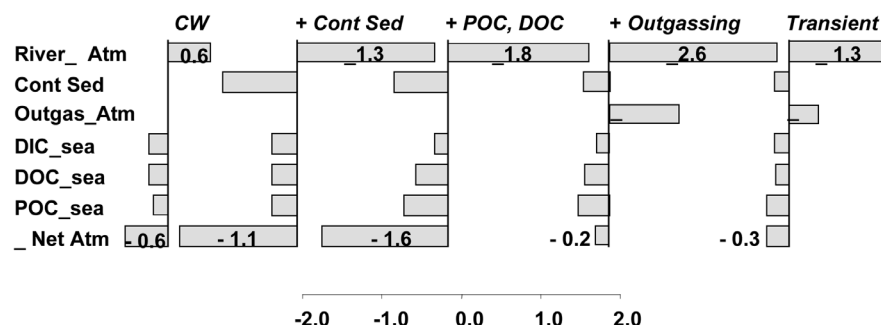
### Overall Fluvial System–Atmosphere Exchange: Alternative Scenarios

Considerable uncertainty remains in the assessment of the carbon cycle of fluvial systems, including the magnitude of fluxes, how to include processes not previously considered, and delineation of anthropogenic and natural processes, all with an explicit recognition of geography. To assess the implications of each, the earlier discussions are summarized here via five “scenarios” (Figure 17.2).

The bulk transfer of atmospheric C through the land to fluvial systems (assuming a steady-state summation of downstream processes, in a non-steady-state environment) ranges from about  $0.6 \text{ PgC y}^{-1}$  (conventional wisdom) to  $2.6 \text{ PgC y}^{-1}$  (+ outgassing). Continental sedimentation results in a significant sink, but that sink is reduced with  $\text{CO}_2$  outgassing (because of the way the sedimentation was computed). The inclusion of continental sedimentation, and then the larger export of OM to the sea (about twice conventional assumptions, under + POC, DOC), yields net sinks of atmospheric  $\text{CO}_2$  of up to  $1.6 \text{ PgC y}^{-1}$ . If outgassing is included, however, then the fluvial net sink is reduced to  $0.2 \text{ PgC y}^{-1}$ . Although partitioning the total fluvial fluxes into natural conditions and anthropogenic transients is problematic at best, substantial evidence suggests that the mobilization of sediments has dramatic increased. While much of this material is captured in reservoirs, it is reasonable to expect that a considerable amount escapes to the sea (especially in non-deltaic regions with steep slopes and few dams).

Summarizing all the components of the riverine carbon cycle, several images emerge. As a global steady-state aggregate, there appears to be a sink (between continental sedimentation and marine sedimentation and dissolution) on the order of  $1\text{--}1.5 \text{ PgC y}^{-1}$ ,





**Figure 17.2.** Scenarios of the fluxes of carbon through fluvial systems relative to atmospheric CO<sub>2</sub>; including atmosphere to rivers (River  $\Delta$  Atm, as sum of other fluxes), continental sedimentation (Cont Sed, alluvial, reservoirs), outgassing from rivers to atmosphere (Outgas  $\Delta$  Atm), export of DIC to the sea (DIC  $\Delta$  sea, as 50 percent of total DIC, to represent just atmospheric weathering component), DOC export to the sea (DOC  $\Delta$  sea), export of POC to the sea (POC  $\Delta$  sea), and by difference the net exchange with the atmosphere ( $\Delta$  Net Atm). For the purposes here, it is assumed that the export fluxes to the sea constitute a sink, with no return to the atmosphere, on an immediate decadal timescale. Particulate inorganic C is not included (as it is abiotic and does not interact with the other pools). Units are PgC y<sup>-1</sup> (scale at bottom lines up with the axis of each graph at 0). Scenarios are CW (conventional wisdom on DOC and POC export, no continental sedimentation or outgassing), + Cont Sed (adding continental sedimentation to CW), + POC, DOC (adding the higher values for POC and DOC fluxes discussed in the text to + Cont Sed), + Outgassing (adding outgassing to + POC, DOC), and Transient (inferring anthropogenic transients, based on the + Outgassing scenario).

est uncertainty) is in South and Southeast Asia and Oceania. Outgassing is function of both pCO<sub>2</sub> concentrations (driven by in situ oxidation) and surface area of water. It is likely most significant in the humid tropics, particularly during the peak of the wet seasons. The highly canalized temperate areas have less area available. The northern latitudes, particularly with warming, are liable to have significant fluxes.

Overall, more carbon is moving through the river system than previously assumed; however, the exact magnitudes remain uncertain. The degree to which fluvial systems constitute a net source/sink relative to the atmosphere is essentially governed by the interplay between mobilization of materials off the landscape and oxidation of those materials back to the atmosphere. Additional data will be necessary to constrain these magnitudes, and it is necessary to partition the model by geographic zone.

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