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Emissions of Methyl Halides and Methane from Rice Paddies

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Methyl halide gases are important sources of atmospheric inorganic halogen compounds, which in turn are central reactants in many stratospheric and tropospheric chemical processes. By observing emissions of methyl chloride, methyl bromide, and methyl iodide from flooded California rice fields, we estimate the impact of rice agriculture on the atmospheric budgets of these gases. Factors influencing methyl halide emissions are stage of rice growth, soil organic content, halide concentrations, and field-water management. Extrapolating our data implies that about 1 percent of atmospheric methyl bromide and 5 percent of methyl iodide arise from rice fields worldwide. Unplanted flooded fields emit as much methyl chloride as planted, flooded rice fields.

Chlorine and bromine catalytic reactions are critical components of global ozone loss (1, 2). Inorganic halogen compounds may also have an appreciable impact on tropospheric and mid-latitude lower stratospheric O_x, NO_x, and HO_x cycles (3–6). The Montreal Protocol regulates many anthropogenic compounds, including chlorofluorocarbons, halons, and methyl bromide, because they provide the majority of inorganic free halogen radicals to the stratosphere. Methyl iodide (CH₃I) may replace methyl bromide (CH₃Br) as it is phased out of use as a fumigant (7). As the anthropogenic contributions to emissions of these gases decrease, a steady state will be reached in which biogenic and other natural inputs become the sole contributors to the global halogen radical burden.

Methyl halides also provide halogen radicals to the troposphere. Currently, the global atmospheric burdens for methyl chloride (CH₃Cl), methyl bromide, and methyl iodide are 3.8 Tg (10¹² g), 150 Gg (10⁹ g), and 1.8 Tg (8–10). The tropospheric lifetimes of CH₃Br and CH₃Cl allow significant fluxes to the stratosphere, whereas the shorter lived CH₃I is fully consumed in the troposphere. Although the magnitudes of various sinks of atmospheric CH₃Cl and CH₃Br are reasonably well known, the individual source magnitudes remain uncertain (8, 11–13). Known methyl chloride sinks exceed known sources by ~1.5 Tg/year, whereas CH₃Br sinks exceed sources by nearly 45 Gg/year (11). Methyl iodide requires a more accurate understanding of both source and sink terms to fully explain its atmospheric budget (14).

Methyl halides are emitted by terrestrial plants and rice grown in the laboratory (13, 14).

Rice paddy methane, a major contributor to the global methane budget, is generated through microbial metabolic processes in the soil and transported through the rice plant to the atmosphere. Methane emissions are dependent on soil organic matter, stage of rice growth, and rice cultivar (15, 16). It is conceivable that methyl halide generation may occur as a metabolic by-product of methane generation. To investigate the mechanisms and magnitudes of their fluxes, we measured methane and methyl halide emissions from two rice plots in Maxwell, California, over the 1998 and 1999 growing seasons. Sample collection, preparation, and analysis followed prescribed methods (8). Fluxes from previous Texas and Japan rice paddy seasons were also measured. The observed emissions were extrapolated to a global scale to assess the impact of rice agriculture on methyl halide budgets.

Rice progresses through four specific life cycle stages (17). Our sampling frequency was sufficient to capture methyl halide fluxes from all four distinct growth stages (18). These fluxes had a noticeable impact on background methyl halide concentrations (19). The data show consistent seasonal emissions of methyl iodide for both Maxwell seasons (Figs. 1 and 2). Methyl iodide appears to have maximum emissions during the vegetative phase, followed by a rapid decrease that starts near booting and continues throughout the season. The decrease in emissions may be interrupted during heading and flowering, as shown by slight increases near day 85 for 1998 and day 105 for 1999. Reproductive stage fluxes in Texas were twice to three times those of Maxwell, whereas late ripening stage emissions were nearly double that of these studies. The Japan 1997 and 1998 ripening stages also show fluxes equivalent to or nearly double that of the Maxwell plots (8). Data from late in the 1997 season at Maxwell are indistinguishable from late season data from 1998 and 1999. Maxwell con-

trol plots emitted negligible (~1.5%) fluxes of methyl iodide (Table 1).

Methyl bromide emissions from the Maxwell study plots during 1998 and 1999 were similar in seasonal pattern and integrated seasonal flux (Figs. 1 and 2 and Table 1). Emissions increased through tillering and appeared to peak during the reproductive stage of rice growth with significant week-to-week variability. Emissions decreased after flowering, in some instances showing methyl bromide uptake. In Texas, the Lemont cultivar during heading may emit nearly five times what was measured at Maxwell, whereas the Mars cultivar shows emissions similar to those seen at Maxwell (8). During ripening, both Lemont and Mars cultivars show fluxes similar to those of this study. The data from Japan 1997 and 1998 and from Maxwell 1997 are similar to the emissions seen at Maxwell 1998 and 1999. Interestingly, the Japan data suggest proportionality between methyl bromide emissions and soil bromide content (8, 20). Maxwell control plots emitted about 10 to 20% the planted flux of methyl bromide (Table 1).

Methyl chloride values appear to be unaffected by stage of growth, which suggests a different mechanism for methyl chloride generation within the rice field (Figs. 1 and 2). Similar to methyl bromide, the data from Japan suggest proportionality between soil chloride content and methyl chloride emissions (8, 20). There may be a slight trend toward larger emissions later in the season, but this is difficult to detect because of the variability of the methyl chloride data. Methyl chloride emissions from the unplanted controls are indistinguishable from the planted plots (Figs. 1 and 2 and Table 1). The similar pattern and magnitude (3.0 × 10⁻⁵ to 4.0 × 10⁻⁵ g m⁻² day⁻¹) of CH₃Cl flux observed in the control plots suggest that CH₃Cl production is independent of methane, CH₃Br, and CH₃I production. Other flooded environments have shown methyl chloride emissions ranging from 1.7 × 10⁻⁶ to 2.9 × 10⁻² g m⁻² day⁻¹ (12, 21).

Methane emissions increased throughout each season, especially after tillering. These results are similar to those from Maxwell 1997 and from previous studies showing that increased methane flux is mainly due to increased soil methane concentration and rice stem diameter (15, 22). The emission profile of methane throughout the season appears to differ from that of each methyl halide, suggesting differences in the pathway of production or the rate of soil consumption for each gas. Analyzing correlations with methane reveals *R* values that are negative for CH₃I, 0.2 for CH₃Cl (*n* = 52), and 0.45 for CH₃Br (*n* = 52). The latter correlation may be significant.

The spike in methane flux at the end of the 1998 season is due to a sudden release when the soil has dried enough to allow methane to escape directly to the atmosphere (17, 23,

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24). No such pattern was observed for any methyl halide, hence methyl halides are not concentrated in the soil in the same manner as methane. This suggests that methyl halides are not produced in the soil by the same anaerobic reactions that produce methane, or that they are consumed more rapidly than they are created.

The amount of available organic matter in

the straw-incorporated (SI) plots was about 50% greater than in the burnt straw plots (15). This study showed no significant correlation between available organic matter and methyl chloride production. In the SI plots, methyl iodide and methyl bromide generally have larger daily fluxes (Figs. 1 and 2), and integrated seasonal emissions (Table 1) are consistently larger. For the 1998 and 1999 Maxwell seasons,

integrated emissions for methyl bromide were ~60% larger for the SI fields. Methyl iodide integrated emissions for the SI fields were equivalent to 87% and 221% of burnt straw field emissions in 1998 and 1999, respectively (Table 1). These results may be influenced by the drainage and reflooding of the plots during 1998, as discussed below. Although variability within the plot makes an accurate assessment of

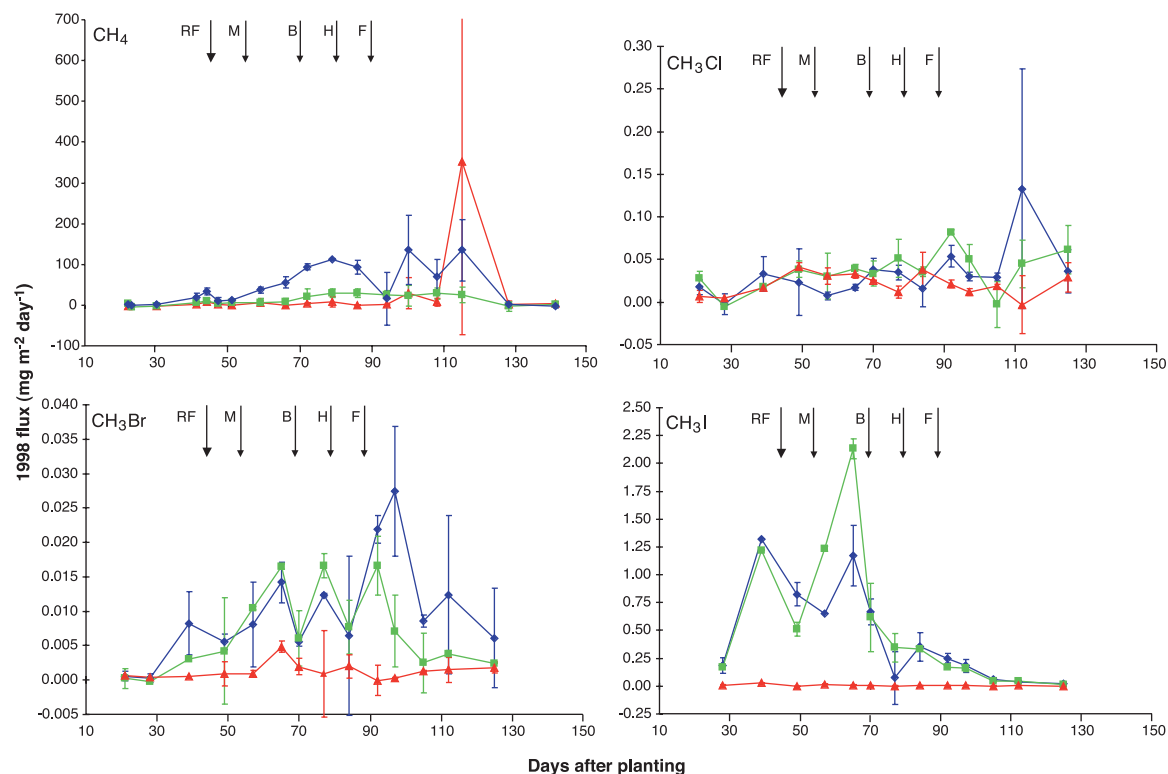


Fig. 1. Maxwell, California, averaged weekly fluxes during 1998 for methane, methyl chloride, methyl bromide, and methyl iodide. Arrows indicate maximum tillering (M, 55 DAS), booting (B, 70 DAS), heading (H, 80 DAS), flowering (F, 90 DAS), and the reflooding date (RF, 45 DAS). The flux for all gases is shown; note differing scales of emission for each gas. Symbols: \blacklozenge , straw-incorporated plots; \blacksquare , burnt straw plots; \blacktriangle , controls. Error bars show one standard deviation.

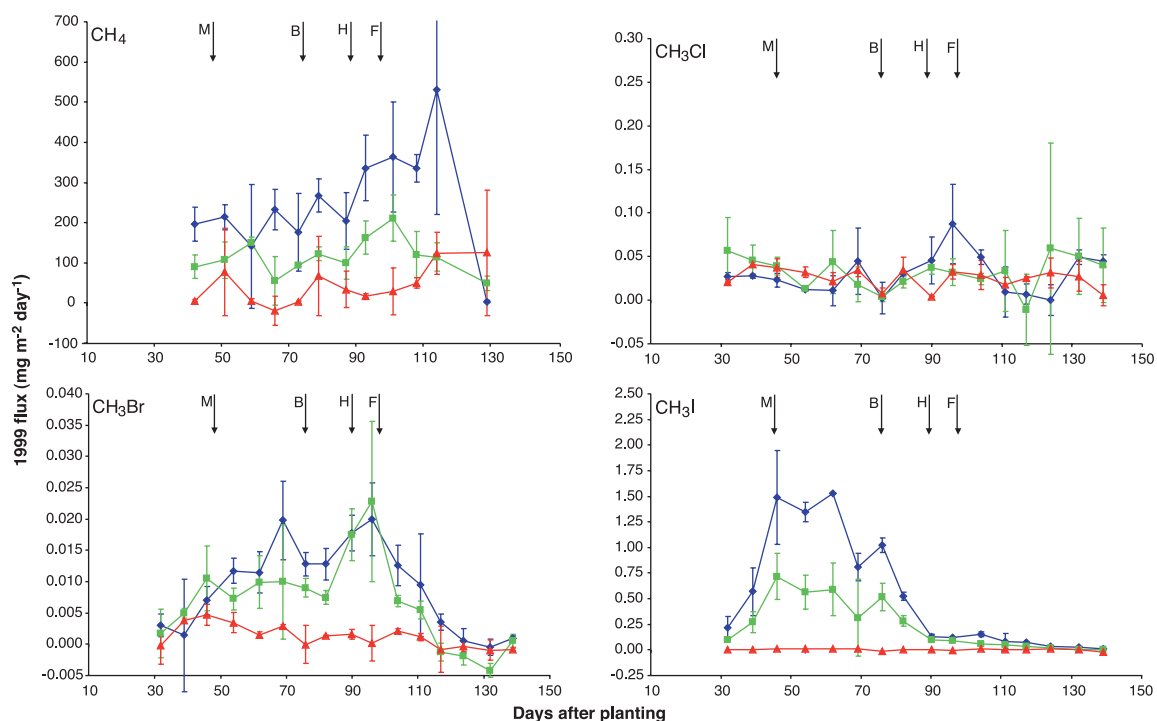


Fig. 2. Maxwell, California, averaged weekly fluxes during 1999 for methane, methyl chloride, methyl bromide, and methyl iodide. Arrows indicate maximum tillering (M, 47 DAS), booting (B, 75 DAS), heading (H, 89 DAS), and flowering (F, 97 DAS). The flux for all gases is shown; note differing scales of emission for each gas. Symbols: \blacklozenge , straw-incorporated plots; \blacksquare , burnt straw plots; \blacktriangle , controls. Error bars show one standard deviation.

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the impact of available organic matter difficult, the differences in CH₃Br and CH₃I emissions between the SI and burnt straw fields are compelling. SI plots had much larger methane production than burnt straw plots (380% and 230% for 1998 and 1999, respectively; Table 1), similar to previous results (15). The impact of soil organic matter on methane emissions is much larger than its observed impact on CH₃Br or CH₃I emissions, again suggesting that methyl halides are not produced in the soil by the same biological processes that produce methane.

During 1998, the plots were drained 41 days after seeding (DAS) and reflooded at 44 DAS to apply herbicides. Emissions of methane and methyl iodide were depressed immediately after herbicide application and reflooding. During 1999, when the field remained flooded, both methane and methyl iodide either maintained or increased their emissions during the same growth period. The week-to-week variability of methyl bromide and methyl chloride emissions during the vegetative stage of rice growth for both seasons makes it difficult to draw distinctions between flooded versus reflooded fields.

Concentrations of halogens in soils decrease from chlorine to bromine to iodine (8). Methyl iodide emissions exceeded those of methyl chloride and methyl bromide, despite the general decreasing trend in ion concentration. On a per-weight basis, assuming a well-leached soil halide concentration (8), the expected ion-driven methyl halide gas emission ratio would be 48:2:1 for methyl chloride to methyl bromide to methyl iodide. The average fluxes we observed were in the ratio 8:2:100. Emission of methyl iodide may provide an important transport mechanism for iodine between the soil and the atmosphere (8).

These data suggest that iodide is preferentially selected by an active biological process within the rice field with decreasing preference for bromide, then chloride. An earlier study has shown a similar iodine preference in *Brassicacea oleracea* (13). Other studies have shown either a marked metabolic preference for chloride ion over bromide and iodide or larger methyl chloride emissions despite enzymatic preference for iodide or bromide ions (25, 26). It seems likely that within rice fields there are at least two separate enzymatic pathways where either chlo-

ride or iodide is the preferred substrate. Our data indicate that the dominant process selects for iodine.

If we estimate fluxes from worldwide rice growing by extrapolating over cultivated area, we obtain global flux estimates for methyl chloride, methyl bromide, and methyl iodide of 5.8, 1.3, and 71 Gg/year, respectively (24, 27). The uncertainties for these estimates are difficult to calculate at this time and will depend on variance within rice cultivars and field parameters. This study suggests that worldwide rice production is responsible for ~1% of atmospheric methyl bromide and 4% of atmospheric methyl iodide. Methyl iodide emissions from rice paddies provide a sizable terrestrial source to the global budget. The values are expected to be conservative when Maxwell emissions are compared against previous data from Texas and Japan. The flux of methane from the Maxwell rice paddies is consistent with previous studies that have shown rice paddies to be a major contributor to the global methane budget (15, 28). Harvest yields from our studied plots are well within the standard deviation of yield for other harvested plots at Maxwell, suggesting only minor perturbations to the natural environment of the rice. Growth stage of the rice plant, soil organic content, and flooding events seem to have large effects on methane, methyl bromide, and methyl iodide emissions. Methyl chloride appears to be emitted through the paddy environment itself, not necessarily through rice-mediated pathways (12, 21, 29).

There are several potential explanations for the differences between the Maxwell emissions and our previous studies: The composition and level of microbial populations may shift, soil halide concentrations may be very different, temperature and light levels may differ, and rice cultivar type may change from field to field (11, 13). The global estimate we have provided will necessarily need to be adjusted as further data are provided for different regimes. Our estimates are based entirely on the 1998 and 1999 Maxwell, California, seasons; our few samples from other years and locations suggest that emissions may be significantly different elsewhere (8). Finally, although the evidence we have presented indicates that production and consumption processes for methane, methyl chloride, methyl bromide, and methyl iodide

may differ from one field to another, it is still uncertain whether the emitted methyl halides are produced in the soils or in the plants themselves.

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17. The first stage is the active vegetative stage during which the plant grows rapidly, both in height and tiller (shoot) number. The active vegetative stage ends at the point of maximum tillering, determined by the period of maximal height gain rate. The lag vegetative stage follows during which the rice continues to grow, albeit at a slower rate than previously. This stage ends with panicle initiation. The panicle is the specific stem on which the grains develop and mature, and at initiation is merely a transparent 2.5-cm-long structure within the stem. Booting follows as the panicle grows in length and diameter, widening the stem as it pushes its way through. Heading occurs as the panicle pushes out of the stem and the panicle is released out of the protective flag leaf. Panicle initiation, booting, and heading occur during the reproductive stage. Flowering occurs quickly after heading, after which the rice expends most of its resources to ripen the grain. Flowering and ripening of the grain constitute the ripening stage of rice growth. A flash of methane may be seen at the end of the ripening stage during field drainage. This process lasts for a very short period, several hours to days, and is difficult to detect without nearly continuous flux measurements. Up to 10% of the methane released to the atmosphere may occur at this time. The 1998 Maxwell rice growth stages were as follows: maximum tillering, 55 DAS; booting, 70 DAS; heading, 80 DAS; flowering, 90 DAS; and harvest on 130 DAS. The stages for the 1999 Maxwell season were as follows: maximum tillering, 47 DAS; booting, 75 DAS; heading, 89 DAS; flowering, 97 DAS; and harvest on 145 DAS [Rice Information Cooperative Effort, *RICE Production Manual* (University of the Philippines, College of Agriculture, in cooperation with the International Rice Research Institute, 1967), pp. 21–25].
18. The data shown are the fluxes averaged from the chamber placements in each plot. Flux data for the

Table 1. Seasonal flux from Maxwell fields, 1998 and 1999. Standard deviations are in parentheses.

	Seasonal flux, 1998 (mg/m ²)			Seasonal flux, 1999 (mg/m ²)		
	Straw-incorporated	Burnt straw	Control plots	Straw-incorporated	Burnt straw	Control plots
Methane	6100 (400)	1600 (200)	800 (500)	27,000 (9400)	12,000 (3800)	5300 (5900)
Methyl chloride	4.1 (3.0)	4.2 (1.7)	2.6 (1.1)	3.7 (1.7)	3.9 (3.3)	3.0 (0.9)
Methyl bromide	1.1 (0.5)	0.7 (0.2)	0.1 (0.1)	1.1 (0.4)	0.8 (0.4)	0.2 (0.2)
Methyl iodide	48.8 (8.4)	56.2 (4.6)	0.7 (0.4)	62.0 (11.6)	28.1 (10.1)	0.5 (0.9)

- control chambers from both fields were averaged together in the same fashion. All emissions data are "net emissions," as both production and consumption of methyl halides may occur in the sample chamber.
19. Methyl bromide and methyl iodide background concentrations, taken at 1 meter height, showed marked increases from early season concentrations of 12 and 2 pptv (parts per trillion by volume). Methyl bromide concentrations of 20 to 50 pptv were observed for the last 2 months of the season, and a prolonged increase in methyl iodide concentrations (30 to 200 pptv) was measured during the early to midseason.
 20. The bromide content in the Japanese fields is 0.7 and 1.1 mg/kg dry weight, respectively, and emissions from these fields are 3.7×10^{-6} and 5.8×10^{-6} g m⁻² day⁻¹ for the listed bromide concentrations. The chloride ion concentration in the Japanese fields was 120 to 450 mg/kg dry weight. Observed emissions of methyl chloride in Japan were 5.8×10^{-5} and 1.5×10^{-4} g m⁻² day⁻¹.
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 29. Methyl chloride emissions from control plots were

very similar to emissions from planted plots. When extrapolating using global rice-field area, control plot emissions are equivalent to 4.1 Gg/year. When the global wetland area of 3.56×10^{12} m² is used, extrapolated emissions of methyl chloride are 10.1 Gg/year. These emissions may be due to a process similar to that responsible for emissions of methyl chloride in fens and salt marshes.

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Early Permian Bipedal Reptile

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A 290-million-year-old reptilian skeleton from the Lower Permian (Asselian) of Germany provides evidence of abilities for cursorial bipedal locomotion, employing a parasagittal digitigrade posture. The skeleton is of a small bolosaurid, *Eudibamus cursoris*, gen. et sp. nov., and confirms the widespread distribution of Bolosauridae across Laurasia during this early stage of amniote evolution. *E. cursoris* is the oldest known representative of Parareptilia, a major clade of reptiles.

During the Late Pennsylvanian–Early Permian, terrestrial tetrapods diversified greatly and adapted to a broad array of herbivorous and predatory modes of life (1). Yet even the most advanced terrestrial amniotes of this time exhibit skeletal features that are indicative of awkward, relatively slow locomotion, such as massive girdles, short stocky limbs, and a "sprawling" posture and gait. We report here on the discovery of a small, nearly complete skeleton of an Early Permian reptile whose skeletal anatomy indicates a capability to attain relatively high speeds during bipedal and quadrupedal locomotion using a parasagittal gait and digitigrade stance. This reptile precedes bipedal parasagittal archosaurs (including dinosaurs) by at least 60 million years.

E. cursoris gen. et sp. nov. (2) is part of a recently collected diverse assemblage of fully terrestrial tetrapods from the Lower Permian of central Germany, near the village of Tambach-Dietharz, Thuringia (3). *E. cursoris* is referred

to the rare, poorly understood taxon Bolosauridae, which includes *Bolosaurus striatus* from the Lower Permian of Texas (4), *Belebey vegrandis* and *B. maximi* from the Upper Permian of Russia (5), and evidence of bolosaurid remains from the Upper Permian of China (6). Until now, however, bolosaurids were known almost exclusively by their skulls because postcrania were limited to a few vertebrae and incomplete limb and girdle elements of *Bolosaurus* (4).

The holotype and only known specimen of *E. cursoris* consists of a nearly complete skeleton, with only a few elements of the skull, forelimb, and the tip of the tail being either missing or incompletely represented (Fig. 1). The skull is partially disarticulated but clearly possesses several bolosaurid characters. Most prominent is a long, low, temporal opening bordered dorsally by the jugal and squamosal and ventrally by the quadratojugal. Only a small remnant of the latter bone is preserved, projecting anteriorly from the ventral margin of the left squamosal. A long, narrow, boomerang-shaped postfrontal wraps around the posterodorsal margin of the orbit. As in other bolosaurids, the palate is devoid of teeth, and the greatly reduced transverse flange of the pterygoid lies in the same plane as the palate proper. In the lower jaw, as in other bolosaurids, the dentary and surangular form a high, broad-based coronoid process, and a medial lamina of the dentary extends nearly to the ventral margin

of the jaw and is covered by a long anterior extension of the prearticular. The upper and lower marginal dentition exhibits the highly unusual bolosaurid structure of bulbous occluding teeth that is considered indicative of high-fiber herbivory (7).

The postcranial skeleton of *E. cursoris* has 26 presacral vertebrae with broadly swollen neural arches and very slender, short ribs. The unusual length of the tail, which includes 55 preserved vertebrae, is due to an elongation of the midregional and posterior elements. The appendicular skeleton has a combination of proportional and structural features that is strongly persuasive of an ability to run not only bipedally but with the hindlimbs held in a nearly erect vertical position and swung in a pendulumlike parasagittal arc, with the pes assuming a digitigrade posture. This form of locomotion is unique among Paleozoic tetrapods. It has been generally accepted that the first appearance of the initial stages of parasagittal limb orientation occurred in the Early Triassic archosaurian antecedents of dinosaurs and crocodylians, whereas the achievement of a fully erect limb posture and parasagittal bipedalism is first documented in the Late Triassic saurischian and ornithischian dinosaurs (8). On the other hand, the earliest appearance of bipedalism without the abandonment of the primitive "sprawling" posture and gait may have occurred in the Late Permian lepidosaurian reptiles (9) and was retained or independently acquired in many modern lizard lineages (10–13). Thus, *Eudibamus* was apparently the earliest known tetrapod capable not only of facultative bipedalism but also of using its hindlimbs primarily in a parasagittal plane with the pes in a digitigrade posture during rapid locomotion (Fig. 2).

The evidence of bipedalism in *Eudibamus* is based principally on specialized skeletal proportions that are not only associated with facultatively bipedal locomotion in modern lizards (10–12, 14, 15) but also characterize fully bipedal dinosaurs that used a parasagittal gait (16). Most compelling are the rela-

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Editor's Summary

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