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Interfield and intrafield variability of methyl halide emissions from rice paddies

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[1] Methyl halide gases are important sources of atmospheric inorganic halogen radicals. We measured methyl halide emissions from three rice fields over two full growing seasons. Rice paddy emissions of methyl chloride, methyl bromide and methyl iodide are insignificant until field flooding. Rice growth stage determines methyl bromide and methyl iodide emissions while methyl chloride emissions are comparable between planted and unplanted plots. Houston, Texas, and Maxwell, California, field integrated seasonal fluxes of methyl chloride, methyl bromide and methyl iodide are consistent (values range from 2.3 to 3.9, 0.8 to 1.1, and 28.1 to 62.0 mg m^{-2}, respectively) despite differences in multiple field parameters. We also examined field emission variability using 12 chamber placements. Methyl bromide and methyl iodide emissions within homogenous rice paddies require at least three replicates to determine field mean fluxes within 20%, and for methyl chloride emissions, over 10 replications per field are necessary. *INDEX TERMS:* 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 1615 Global Change: Biogeochemical processes (4805); 1610 Global Change: Atmosphere (0315, 0325); *KEYWORDS:* methyl halide, methyl chloride, methyl bromide, methyl iodide, emissions, rice

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1. Introduction

[2] Inorganic halogen radicals affect the oxidative capacity of both the stratosphere and the troposphere. In the stratosphere halogen radicals catalytically destroy ozone, creating the well-documented Antarctic ozone hole [*Anderson et al.*, 1991; *Santee et al.*, 1995]. In the troposphere, halogen radicals shift the partitioning of HO_x, NO_x, and O_x species, decreasing the lower atmospheric oxidative capacity and potentially altering the rate of tropospheric hydrocarbon removal [*Vogt et al.*, 1999; *McFiggans et al.*, 2000]. Recent evidence also indicates that biogenic iodine emissions may enhance the production of aerosols [*O'Dowd et al.*, 2002].

[3] A significant source of inorganic chlorine and bromine radicals to the stratosphere is the oxidation of methyl chloride and methyl bromide. Photolysis of methyl iodide and oxidation of methyl chloride and methyl bromide provide a sizable source of inorganic halogen radicals to the troposphere. The relative production of halogen radicals from natural or anthropogenic sources is not yet clear because of our incomplete understanding of methyl halide budgets. Only 60, 75, and at most 40% of the methyl chloride, methyl bromide and methyl iodide sources can be accounted for [*Butler*, 2000].

[4] Recently, terrestrial plants and ecosystems have been marked as potentially significant emitters of methyl halides. Rapeseed and rice agriculture alone may together produce nearly 10 Gg/yr of methyl bromide, nearly 5% of the annual global budget [*Gan et al.*, 1998; *Redeker et al.*, 2000]. These estimates rely on extrapolations that assume field and plant homogeneity. Field studies have shown that even uniform ecosystems, such as tundra and agricultural fields, may have site-to-site variability of gas emissions on the order of 50 to 150% ((CH_4) [*Whalen and Reeburgh*, 1988]; (N_2O) [*Matthias et al.*, 1980]). Whereas agricultural ecosystems, specifically rice paddies, are likely to be more homogenous than most natural systems [*Sass et al.*, 2002], they have shown themselves to be highly variable [*Matthias*

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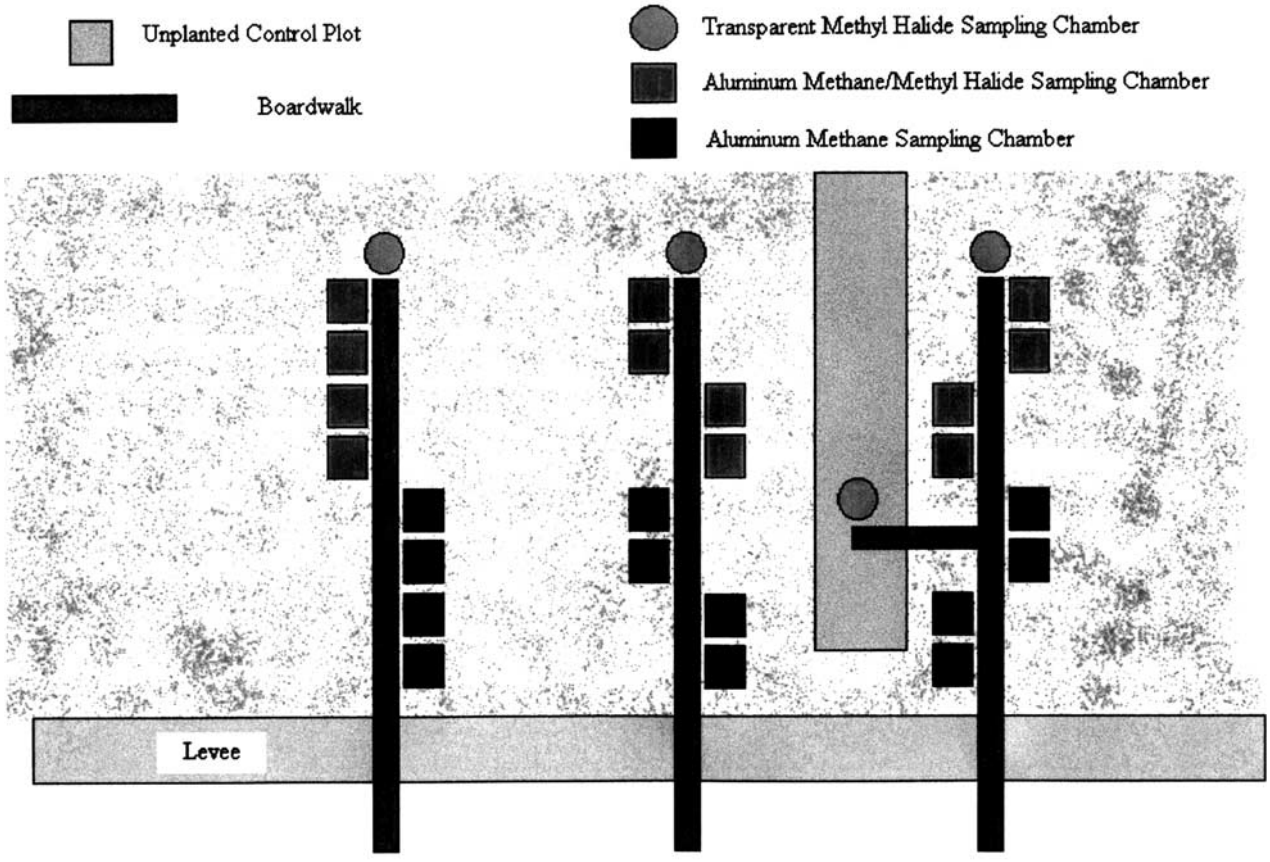

Figure 1. Experimental site description for Houston, Texas, 2000. Diagram is not to scale. Boardwalks are approximately 18 m apart and 20 m long. Opaque square chambers are at least 3 m from the levee. Note control chambers placed in an unplanted portion of the rice paddy similar to the planted chambers.

et al., 1980; *Yagi and Minami*, 1993]. Our previous measurements of methyl halide emissions in rice paddies show site-to-site field variability on the order of 10 to 70% [*Redeker et al.*, 2000]. To extrapolate fluxes from individual field studies to regional and global scales, it is necessary to assess the variability of methyl halide emissions within the field. Further, the impact of cultivar, soil carbon, soil halide, field water management, and temperature on methyl halide emissions must be quantified and integrated into regional and global extrapolations.

[5] In summer of 2000 we studied a field near Houston, Texas, using the same methodology that we used in a previous field campaign in Maxwell, California, during 1999. Our work in Maxwell, California, during 1998 and 1999 showed year-to-year variability of seasonally integrated methyl halide emissions of less than 15% [*Redeker et al.*, 2000]. Here we compare the methyl halide emissions from the seasonal studies in Maxwell, California, during 1999 and Houston, Texas, during 2000. During the 2000 field season we also measured intrafield variability in methyl halide emissions.

2. Methods

[6] Two experiments ran concurrently during the 2000 growing season in Houston Texas. Twenty-four square opaque chambers and four UV-visible transparent chambers were placed along three 21.5-m-long transects evenly located within the field. One transect also included a t-section which allowed us to place a chamber over an unplanted, but flooded region of the field (Figure 1). Chambers were placed to avoid field edge effects; the nearest field border was 20 m from any transect, while transects were separated by 18 m. The total field area was 0.7 ha and the experiment covered nearly one sixth of the total field area.

[7] To study and compare seasonal methyl halide emissions from rice paddies, we used UV-visible transparent polycarbonate chambers placed at the apex of each transect. Chamber flux measurement methodology and sample analyses in Houston were performed as described in *Redeker et al.* [2000] to allow maximum compatibility with the previous Maxwell, California, study (Figure 2).

[8] Methyl halide emission variability within the Houston field was studied for a subset of the available 24 square opaque chambers used for the methane variability study (Figure 1). Samples were taken on three midseason dates, into 0.5- and 2-L electropolished canisters through a stainless steel line immersed within an ice-bath, similar to the method described by *Redeker et al.* [2000]. Fluxes were calculated using the methods of *Redeker et al.* [2000]. Concurrent measurements of field methane variability were taken as described by *Sass et al.* [2002].

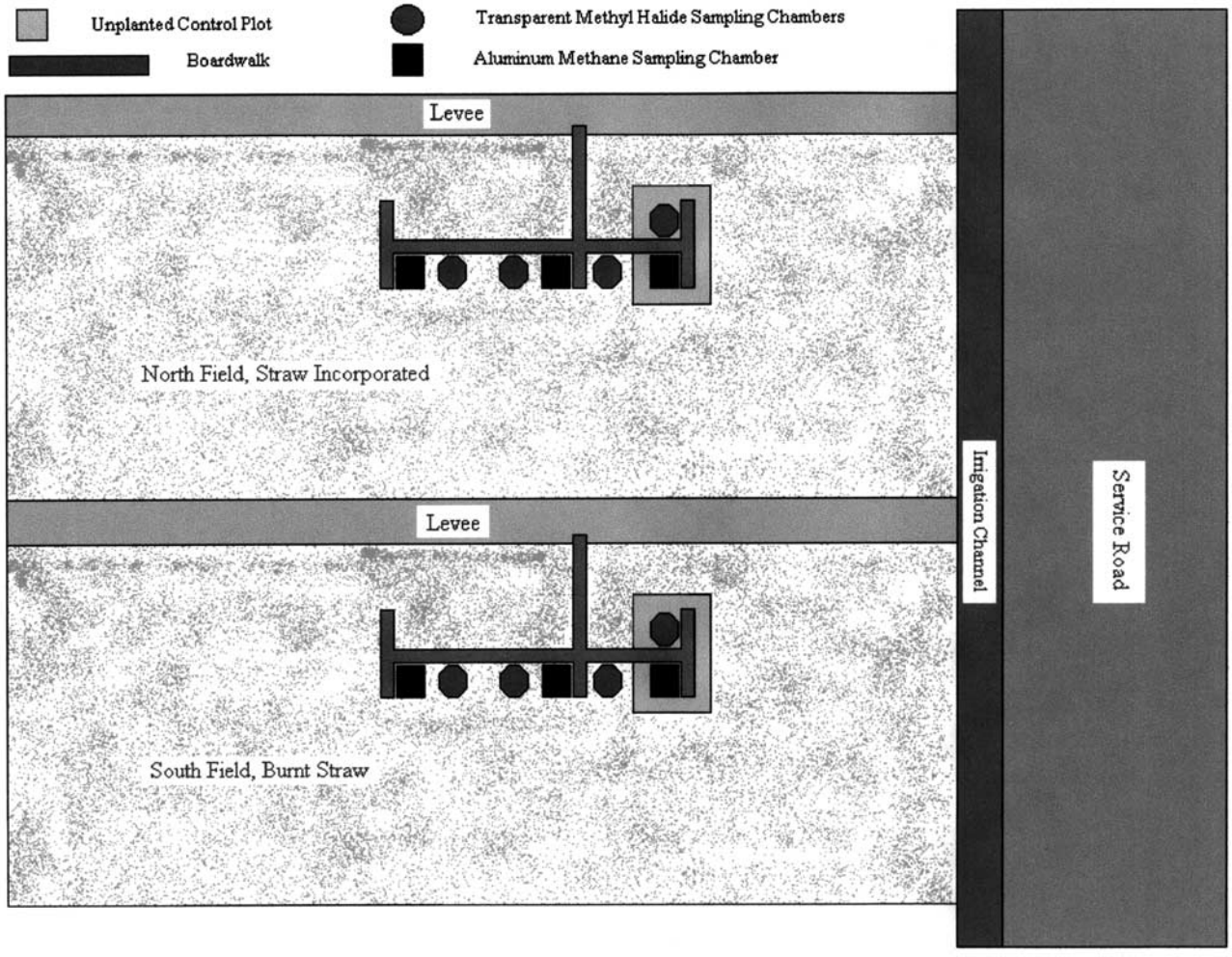

Figure 2. Experimental site description for Maxwell, California, 1999. Diagram is not to scale. Boardwalks extend approximately 8 m into the field, with minimum distance between the levee and any given chamber no less than 4 m. Note the control chambers are placed in an unplanted portion of the rice paddy similar to the planted chambers.

[9] Soil cores were collected from Maxwell, California, during the late ripening stage of the 1998 season. Subsurface samples were collected from Houston prior to field flooding and after harvest. Soil samples were homogenized, then finely ground and analyzed for total carbon content on a Carlo Erba NA-1500 combustion oven [*Verardo et al.*, 1990]. Inorganic soil carbon within the rice paddy soils was less than 5% of the measured total carbon concentrations. Instrumental precision for duplicate analysis of soil carbon was better than 3%. Soil sample variability was somewhat larger than the observed instrumental precision (Table 1).

[10] Soil halide concentrations were determined through multiple extractions of homogenized and sorted soil subsamples. Twenty-five grams of soil were placed in 50 mL of deionized water and shaken every 15 min for an hour. After 1 hour the extract was run through a #3 Whatman filter. The soil sample was then re-extracted a second and third time as described above with 50 mL DI water for each subsequent extraction. Our analysis suggests that single-pass sample extraction efficiencies for soil halides are no better than 72% for chloride (±10%, n = 15) with progressively poorer efficiencies for bromide (62 ± 23%) and iodide (44 ± 30%). We measured both soil iodide and iodate content; to measure iodate, we reduced the extract with ascorbic acid [*Shin et al.*, 1996], then removed the ascorbic acid with ascorbate oxidase. Endpoints of these reactions were determined via spectrophotometric analysis at 520 nm with dichloroindophenol as an indicator.

[11] Aqueous solutions of extracted halides were analyzed with specific ion electrodes in concert with the Gran known addition technique [*Gran*, 1952, *Orion Research, Inc.*, 1991]. The influence of interfering ions is minimized by application of this known-addition specific ion electrode-based method. The inherent instability near the endpoint is also diminished. It allows for halide measurements near their limit of detection; $[Cl^-] = 5 \times 10^{-5}$ M, $[Br^-] = 5 \times 10^{-6}$ M, $[I^-] = 5 \times 10^{-8}$ M. Our use of the Orion/Gran known-addition methodology provided duplicate sample measure-

Table 1. Interfield Variables

	Maxwell, California, 1999	Houston, Texas, 2000
Soil type	fine montmorillonitic, thermic soil; 51% clay	fine montmorillonitic, thermic soil; 51% clay
Total soil carbon content	20.6 gC/kg in straw incorporated 18.4 gC/kg in burnt straw soil carbon variability ±15%	9.0 gC/kg previous straw residue burned soil carbon variability ±15%
Total soil halide content	chloride = 53 mg/kg SI, 59 mg/kg BS bromide = 1.5 mg/kg SI, 1.9 mg/kg BS iodide = 0.06 mg/kg SI, 0.07 mg/kg BS halides highly variable ±16% Cl, ±55% Br, ±19% I	chloride = 102 mg/kg bromide = 4.6 mg/kg iodide = 0.04 mg/kg halides highly variable ±32% Cl, ±70% Br, ±53% I
Field temperature	Average air temperatures for both Maxwell fields were ~31°C. Variations in temperature were seen throughout the season with no specific trend.	Average air temperatures in Houston fields were 32°C. Temperature exhibits an upward trend throughout the season, starting at 29°C and ending near 39°C.
Field water management	Seeds are broadcast over flooded fields. Fields remain flooded throughout the season.	Seeds are drill planted in nonflooded fields. Flooding occurs when rice reaches ~15 cm in height and is maintained thereafter until the end of the season.
Field fertilization	Fields are fertilized with 150 kgN/ha before the seeds are broadcast. No midseason fertilization occurs.	Fields are fertilized before planting and several times during the season, with total fertilization equal to 230 kgN/ha. Midseason application is by airplane broadcast.

ments and known standard calibrations that indicate better than 10% precision in the concentration ranges observed.

3. Results and Discussion

3.1. Interfield Variability

[12] In our previous studies in Maxwell, California, the determining factor in methyl bromide and methyl iodide emissions appeared to be the growth stage of the rice plant. Methyl chloride appeared to be generated by the paddy environment itself and was not correlated with methane, methyl bromide or methyl iodide emissions [*Redeker et al.*, 2000]. Other factors that were potentially related to methyl halide emissions included field water management, soil carbon content, soil halide content and rice cultivar. Our study in Houston, Texas, incorporated changes in all of these parameters relative to previous studies in Maxwell, California (Tables 1 and 2).

3.1.1. Rice Growth Stage

[13] Emission rates of methane, methyl bromide and methyl iodide were dominated by growth stage of the rice plant (Figure 3), easily overpowering differences in rice cultivar, soil carbon content, and soil halide content. Methane emissions were minimal at early sampling dates and increased until midseason, at which point they reached a plateau or increased until field drainage. Methyl bromide emissions increased from early season until panicle initiation, the beginning of the reproductive stage. During the reproductive stage (panicle initiation through flowering) emissions fluctuated, with weekly timescales, around the maxima (Figure 3). After the reproductive stage, methyl bromide emissions decreased rapidly until harvest. Unplanted control plots were responsible for less than 10% of the methyl bromide flux observed from planted chambers. Methyl iodide emissions increased rapidly during the tillering phase with maximum emissions occurring during the period of maximum growth (in height) of the rice plant. Emissions of methyl iodide tended to maximize before panicle initiation and were maintained through heading after which they exponentially decreased. There appeared to have been a late season increase in emissions during flowering, but this increase was small compared to early season fluxes. The control chamber emissions were negligible (<1%) when compared to the measured methyl iodide flux from planted chambers. Methyl chloride emissions remained constant, between 0.02 and 0.08 mg m^{-2} day^{-1}, in both planted and unplanted plots from early season until field drainage (Figure 3), supporting the hypothesis presented by *Redeker et al.* [2000] that methyl chloride emissions are driven by processes different than those driving methyl bromide and methyl iodide emissions. See work by *Redeker et al.* [2000] for a more complete description of rice growth stages.

3.1.2. Field Water Management

[14] After field flooding in Houston, methyl halide emissions, negligible under dry conditions, began to approach fluxes observed in the continuously flooded Maxwell fields (Table 1 and Figure 3). These data from Houston support

Table 2. Cultivar Differences

	Maxwell, California, 1999	Houston, Texas, 2000
Cultivar	M202	cocodrie (cross of cypress/L202/tebonnet)
Rice height, cm		
50 DAS	63 ± 4.5	37 ± 3.9
60 DAS	73 ± 3.2	65 ± 3.2
85 DAS	83 ± 3.8	80 ± 3.0
100 DAS	88 ± 3.5	92 ± 3.3
130 DAS	87 ± 4.5	88 ± 3.5
End of season grain yield[a]	920 ± 65 g m^{-2}	670 ± 90 g m^{-2}
Aboveground biomass yield (w/o grain)[a]	880 ± 180 g m^{-2}	780 ± 140 g m^{-2}

[a]Biomass yields are assumed to be consistent with data reported by *Bossio et al.* [1999] and *Sass et al.* [2002]. Houston aboveground biomass without grain was calculated by subtraction of the panicle weight from the total aboveground biomass.

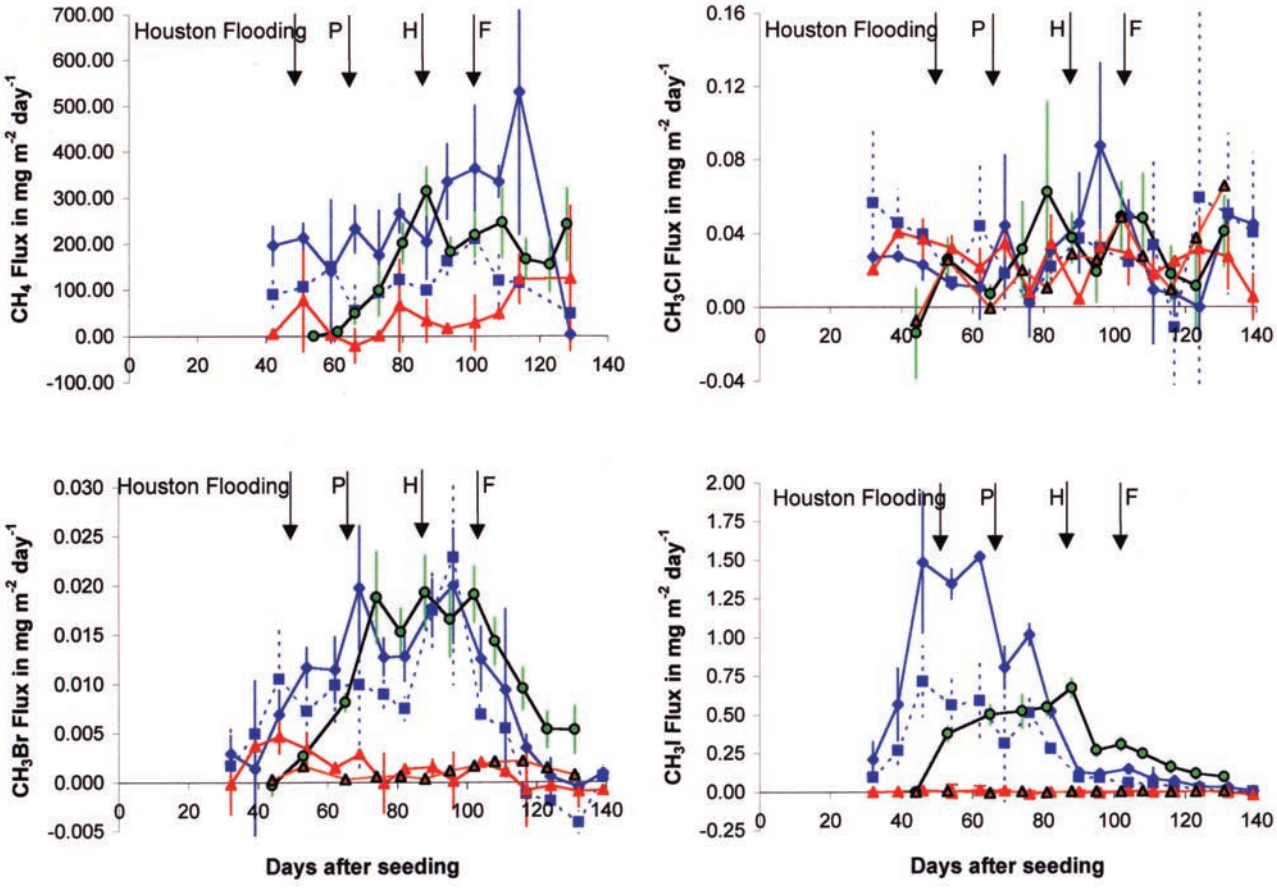

Figure 3. Comparison of Houston, 2000, versus Maxwell, 1999, averaged weekly fluxes. Arrows indicate the Houston flooding date (49 days after seeding, DAS), Panicle initiation (P, ~60 DAS), Heading (H, ~85 DAS) and Flowering (F, ~105 DAS). Symbols are diamonds: Straw Incorporated plots from California; squares: Burnt Straw plots from California; circles: Texas fields; and triangles: controls from both Texas and California. The flux for all gases is shown; note differing scales of emission for each gas. Error bars show 1 standard deviation (n = 3).

our earlier hypothesis, based on midseason field drainage in Maxwell 1998, that methyl halide emissions are strongly dependent on soil pore water saturation [*Redeker et al.*, 2000]. The seasonal patterns of methyl chloride and methyl bromide emissions were unaffected by field water management, but peak methyl iodide emissions were shifted to later in the season. Methyl iodide emissions are most likely to be affected by field flooding practices as they peak in the early season.

3.1.3. Soil Carbon Content

[15] Increased soil organic carbon content increases methane emissions [*Sass*, 1994; *Bossio et al.*, 1999]. While our observed emissions of methyl chloride, methyl bromide, and methyl iodide might indicate a dependence on soil carbon content, the data are inconsistent (Tables 1 and 3). Methyl bromide and methyl iodide emissions in Houston were equivalent to those in Maxwell, California, despite soil carbon levels half those found in Maxwell. The soil carbon-content influence on methyl chloride emissions is too small to be measured, especially when the highly variable nature of the methyl chloride data is considered.

3.1.4. Soil Halide Content

[16] Soil bromide content has been shown to influence methyl bromide emissions from various members of the Brassica family [*Gan et al.*, 1998]. A comparison between Maxwell and Houston soils shows that chloride and bromide ions were relatively enriched in Houston (Table 1). Emissions of methyl chloride, however, decreased from Maxwell to Houston and emissions of methyl bromide remained nearly constant (Figure 3 and Table 3). Soil iodide values were lower in Houston than in Maxwell (Table 1), and we saw comparable emissions of methyl iodide from Houston fields (Figure 3 and Table 3). Any relationship between methyl halide emissions and soil halide content is complicated by high site-to-site variability of soil halide concentrations within the field (Table 1).

3.1.5. Ambient Temperature

[17] Other biogenic volatile organic carbon emissions have been shown to depend strongly on temperature [*Monson et al.*, 1995; *Guenther et al.*, 1995]. A comparison of field temperatures between Houston and Maxwell indicates that ambient field temperatures were similar until near the

Table 3. Seasonal Flux From Maxwell Fields, 1999, and Houston, 2000[a]

	Maxwell, California, 1999			Houston, Texas, 2000	
	Straw Incorporated	Burnt Straw	Control Plots	Planted	Control Plots
Methane	27000 (9400)	12000 (3800)	5300 (5900)	12000 (1700)	
Methyl chloride	3.7 (1.7)	3.9 (3.3)	3.0 (0.9)	2.3 (2.3)	2.1
Methyl Bromide	1.1 (0.4)	0.8 (0.4)	0.2 (0.2)	1.0 (0.2)	0.1
Methyl Iodide	62.0 (11.6)	28.1 (10.1)	0.5 (0.9)	30.8 (4.4)	0.3

[a]Fluxes are in mg/m^2 with standard deviation in parentheses.

Table 4. Linear Methane and Methyl Halide Emission Correlation Coefficients (R), 1998–2000[a]

	1998 Maxwell, California (n = 28)			1999 Maxwell, California (n = 24)			2000 Houston, Texas (n = 33)		
	CH_3Cl	CH_3Br	CH_3I	CH_3Cl	CH_3Br	CH_3I	CH_3Cl	CH_3Br	CH_3I
CH_4	0.32	0.49	−0.25	0.11	0.41	−0.04	0.40	0.49	−0.04
CH_3Cl		0.39	−0.19		0.25	−0.35		0.51	0.30
CH_3Br			0.12			0.15			0.61

[a]Data set size is given in parentheses.

end of the season, when Houston temperatures increased to nearly 39°C while Maxwell temperatures remained constant at 32°C (Figure 4). This temperature difference may have caused higher emissions of methane, methyl bromide and methyl iodide near the end of the Houston season as compared to the late Maxwell season (Figure 3).

3.1.6. Fertilization

[18] The Houston fields were fertilized four times throughout the season (~60kg-N/ha each fertilization) while the Maxwell study sites were fertilized once at the beginning of the season (150kg-N/ha) (Table 1). There was no discernible impact of fertilization on methyl halide and methane emissions although sampling dates tended to be several days after field fertilization and any observable increase in gas fluxes may have already occurred.

3.1.7. Cultivar

[19] Total emission of methane from rice and seasonal methane emission patterns depends upon the cultivar grown [*Sass*, 1994]. The two cultivars grown in Maxwell and Houston were M103 and Cocodrie, respectively. Both varieties are very early maturing, semidwarves, although M103 creates a medium grain and Cocodrie a long grain. Cultivar height, biomass, and grain yields are described in Table 2. Methyl halide emissions are remarkably consistent, suggesting little or no cultivar effect between M103 and Cocodrie, although these effects are difficult to detect during the early season due to the comparatively late flooding date in the Houston fields. The slight, but distinct late season differences (over 2σ) in methyl bromide (>115 DAS) and methyl iodide (>85 DAS) emissions may be due to cultivar effects or to increased temperatures during the Houston late season. These two possibilities remain unclear and need further study.

3.1.8. Component-by-Component Analysis of Variance

[20] Component-by-component correlation analyses were performed to determine if the same processes that drive methane production drive methyl halide production or whether methyl halide emissions are driven by identical metabolic reactions. The results from these analyses were inconsistent within the three years studied (Table 4, n = 28, 24, and 33 in 1998, 1999, and 2000). The methane versus methyl chloride correlation coefficient varied from 0.1 to 0.4 while methane versus methyl iodide emission coefficients were generally poor ($|R| < 0.1$ for 1999 and 2000). In contrast, methane versus methyl bromide emissions maintained consistent correlation coefficients between 0.4 and 0.5. Methyl chloride and methyl bromide values ranged from 0.25 to 0.5, which may suggest a positive correlation. These correlations must be viewed with some caution as methyl chloride versus methyl iodide emissions showed widely different correlation coefficients of −0.2, −0.3 and 0.3 in 1998, 1999, and 2000, respectively (see also the discussion on intrafield variability of methyl chloride emissions, below). Finally, methyl bromide versus methyl iodide

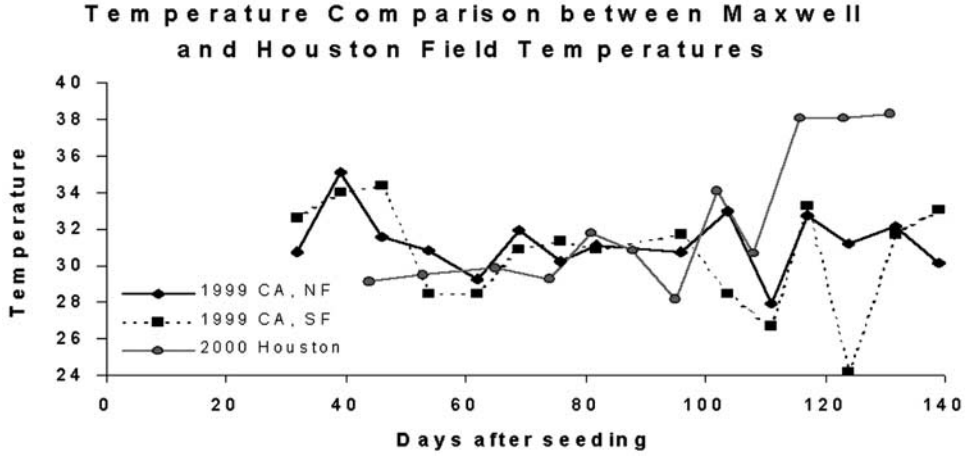

Figure 4. Early afternoon ambient air temperature comparison between Maxwell, 1999, and Houston, 2000, field experiments.

Table 5. Analysis of Chamber Variance With All Possible Two-, Three-, and Four-Chamber Sets[a]

	5/17/00			5/31/00			6/14/00		
	2	3	4	2	3	4	2	3	4
Percent Probability of the Sum of Chambers Lying Within 5% of Measured Mean									
MeCl	1.5	11.8	11.7	3.6	3.6	6.1	7.3	6.7	6.4
MeBr	10.6	13.2	18.8	21.8	33.3	42.7	27.3	34.5	41.5
MeI	13.6	16.8	20.0	43.6	53.9	60.3	23.6	42.4	42.4
Percent Probability of the Sum of Chambers Lying Within 10% of Measured Mean									
MeCl	16.7	21.4	23.6	7.3	7.3	12.1	9.1	9.7	11.8
MeBr	21.2	25.9	36.2	49.1	61.8	72.7	56.4	65.5	75.5
MeI	27.3	31.4	35.2	70.9	83.0	93.0	49.1	66.1	80.0
Percent Probability of the Sum of Chambers Lying Within 20% of Measured Mean									
MeCl	33.3	35.9	45.5	18.2	18.2	25.2	18.2	18.8	22.7
MeBr	45.5	55.9	68.3	80.0	94.5	98.2	89.1	95.2	99.7
MeI	54.5	54.1	65.3	96.4	100.0	100.0	89.1	95.2	99.4
	MeCl			MeBr			MeI		
	2	3	4	2	3	4	2	3	4
5%[b]	4.1	7.4	8.0	19.9	27.0	34.3	27.0	37.7	40.9
10%[b]	11.0	12.8	15.9	42.2	51.1	61.4	49.1	60.2	69.4
20%[b]	23.2	24.3	31.1	71.5	81.9	88.7	80.0	83.1	88.2
95% confidence interval	mean ± 66.80%			mean ± 17.56%			mean ± 16.26%		

[a]Numerals 2, 3, and 4 in heading refer to the number of chambers.
[b]Probability of the average of 2, 3, or 4 chambers lying within the given percent of the measured mean.

emissions demonstrated a broad range, from approximately 0.1 in Maxwell to 0.6 in Houston.

[21] These correlations, which might be construed as indicative that similar processes drive methane and methyl halide production, should be approached with some caution due to the highly variable, nonnormal distribution of emissions in the field, along with the heavy influence on emissions by outside biogenic factors, including when and how the rice paddies are flooded, ambient temperature, etc.

3.2. Intrafield Variability

[22] Just as *Sass et al.* [2002] studied methane flux variability within a Houston rice paddy, we were able to assay the spatial variability of methyl halide emissions. Intrafield variability of methyl halide emissions decreased over the course of the season, in a similar fashion to methane emissions (Table 5 and *Sass et al.* [2002]). As described by *Sass et al.* [2002], the frequency distribution of mean field emissions derived from two-, three-, and four-flux-chamber experiments can be constructed if we assume that the measured sample set (Figure 1) describes the entire field heterogeneity. If we consider all combinations of two, three, and four chambers within the field, the total number of possible combinations within the 12-chamber site are 66, 220, and 495, respectively. As expected, with larger sample size (3 and 4 chambers) comes more possible combinations and a more normal distribution of mean methyl halide emissions (Figure 5).

3.2.1. Precision of Two-, Three-, and Four-Chamber Replications

[23] The probability of a flux calculated from a two-chamber experiment to lie further than 1 standard deviation from the true mean can be as high as 20%. The potential for fluxes calculated from a three-chamber experiment to lie farther than 1 standard deviation from the actual field mean is much smaller (<5%), and with four chambers it is negligible (<1%). Our study shows that the probability of the measured value from a three-chamber experiment to be within 20% of the true field mean for methyl bromide and methyl iodide is close to 55% in the early season but approaches 100% in the late season. Four-chamber studies give similar results, with most accuracy gained in the early season during the highly variable period (Table 5). In contrast, observed rice emissions from a two-chamber study would begin the season near 50% probability of being within 20% of the actual field mean and are only likely to reach 85–90% during the ripening stage. These values suggest that to measure methyl halide emissions accurately from homogenous systems (including rice paddies), three chambers are necessary. Heterogeneous systems will require more than three-chamber replications for accurate estimates of regional fluxes.

3.2.2. Methyl Halide Emission Variability

[24] Methyl chloride variability within the field was consistently larger than the variability shown by methyl bromide and methyl iodide (Table 5). With methyl halide measurements from 12 chambers, the calculated 95% confidence interval is the measured field mean plus or minus 67, 18, and 16% for methyl chloride, methyl bromide, and methyl iodide. The large variability of methyl chloride when compared to methyl bromide and methyl iodide further supports our hypothesis that methyl chloride is created through a different mechanism than methyl bromide and methyl iodide in the rice paddy ecosystem.

3.3. Summary

[25] In comparisons between field studies in Maxwell, California, 1999, and Houston, Texas, 2000, we find that methyl halide emissions are strongly rice-growth stage and field-water management dependent and possibly weakly dependent on cultivar, soil organic carbon content, soil

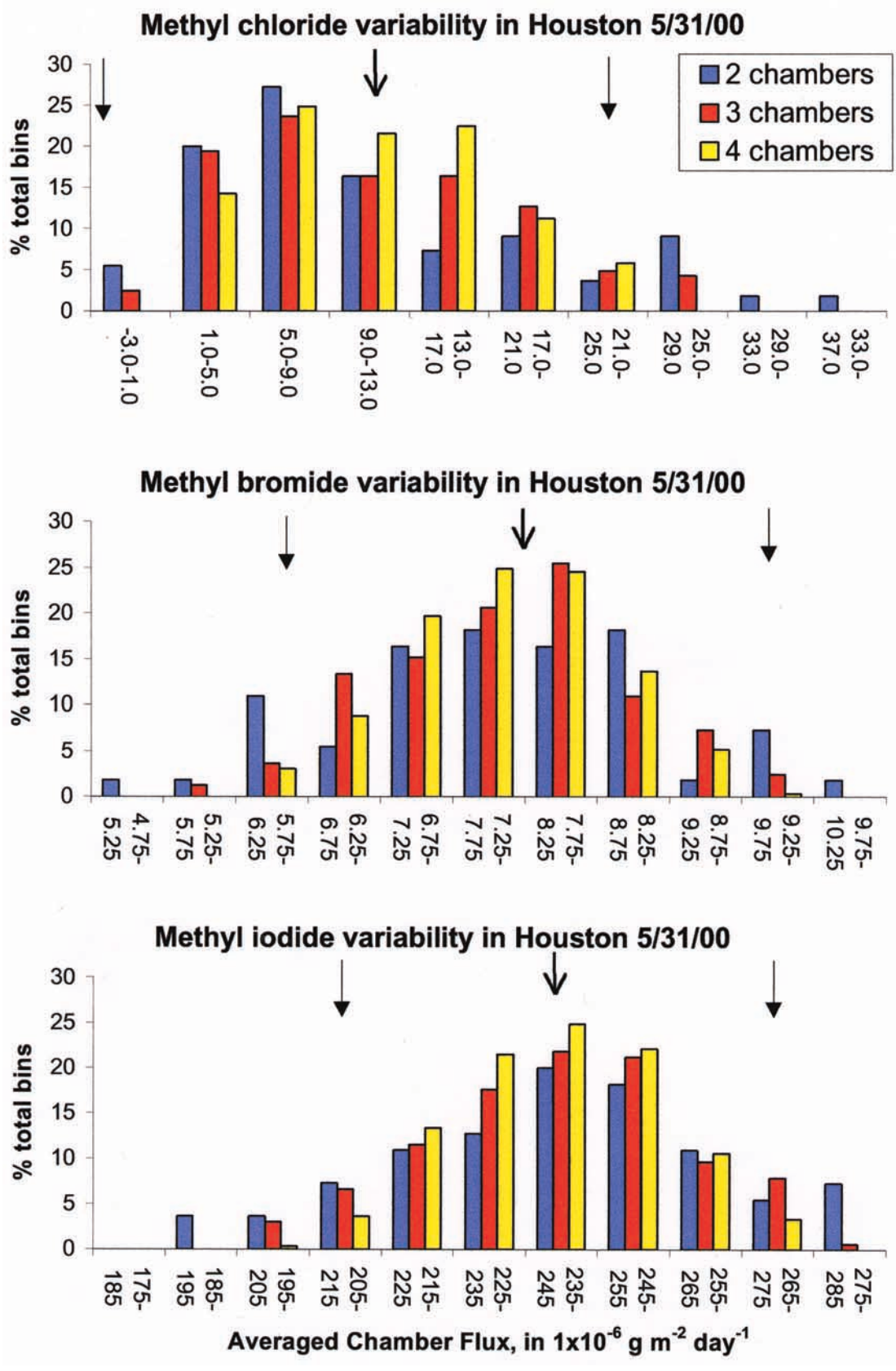

Figure 5. Methyl halide field flux variability data from Houston, Texas, 31 May 2000. Thick, open-point arrows indicate the daily flux average for all bins, while thin, closed-point arrows indicate 1 standard deviation from the mean.

halide content, and ambient field temperature. Field-fertilization practices (nitrogen) appeared to have no influence on methyl halide emissions. Our estimates of methyl halide emissions from rice paddies, based on the three-chamber field studies in Maxwell and Houston, are 3.3, 1.0, and 40.3 mg/m^2 of methyl chloride, methyl bromide, and methyl iodide, respectively, per season. Extrapolating to global rice areas (1.5×10^{12} m^2 (Statistics Norway, Statistical Year-Book 1998, retrieved November 7, 2000, from the World Wide Web at http://www.ssb.no/english/yearbook/1998/tab/t1510002.shtml) we calculate possible ranges of rice paddy emissions to be 3.5 to 5.9, 1.2 to 1.7, and 42 to 93 Gg/yr for methyl chloride, methyl bromide, and methyl iodide based on our three-field campaigns. These ranges may be compared to our previously reported values from *Redeker et al.* [2000] of 5.8, 1.3, and 71 Gg/yr and are equivalent to 0.1 to 0.2, 0.6 to 0.9, and 3 to 5% of the necessary annual global source for each compound. Within 95% confidence, our measured methyl halide field fluxes are within 20% of the actual field mean for methyl bromide and methyl iodide while our observed methyl chloride flux is accurate only within 70%. Further work on the effects of cultivar, soil carbon content, soil halide content, and field water management are necessary to describe regional methyl halide emissions from rice paddies.

[26] Methyl halide emission variability depends on growth stage of the rice, possibly due to heterogeneity of shading and temperature effects from an incomplete rice canopy. Intrafield variability analysis suggests that analyses with four chambers are more accurate when extrapolating to regional and global annual fluxes than those that use two or three chambers. However, three-chamber experiments are capable of capturing the field mean emissions within 20% for methyl bromide and methyl iodide. Extrapolations of methyl halide emissions from other terrestrial systems based on less than three replications and/or inadequate representation of annual growth stages/cycles should be viewed with some caution.

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