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Heat Flux Measurements and Modeling of Malodorous Compounds above an Anaerobic Swine Lagoon

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Abstract The concentration of *p*-cresol and *p*-ethylphenol, two malodorants typical of swine waste, were measured at 0.5 and 1.5 m above a waste treatment lagoon during two separate campaigns encompassing late winter through early spring and late spring through early summer. Concomitant collection of air temperatures, humidities, insolation, and wind speeds, as well as water column temperatures were done so that heat fluxes could be computed using an energy budget method and Bowen ratio estimates. The empirical model that was found to correlate best with variations in malodorant concentrations and gradients above the lagoon had the terms describing evaporation from the lagoon surface and net available energy at the lagoon surface. Emissions were found to be much higher during the cool season than the warm season. This was despite much higher evaporation rates during the warm season. This could be explained by much lower lagoon concentrations of the malodorants in the warm season than in the cool season. Results of this work are being used to determine appropriate models to estimate malodorant emissions from lagoons and devise techniques for the abatement of nuisance emissions.

Keywords *p*-cresol · *p*-ethylphenol · Evaporation · Heat flux · Lagoon · Malodor

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1 Introduction

Waste from concentrated animal feeding operations are typically stored and treated in large earthen lagoons prior to land application of wastes (Richard and Hinrichs 2002). In that these lagoons are open to the atmosphere, there is emission of malodorous compounds that can cause complaints by neighbors and may have adverse environmental affects (Schiffman et al. 2001; Lim et al. 2003).

Of these malodorous compounds, the emission of ammonia has received the most attention both of because of its relative abundance in wastes and due to adverse environmental impacts such as eutrophication of waters (Paerl 1997; Anderson et al. 2002). Relatively high emission rates, and the ability to capture ammonia in acid traps, facilitate its capture and analysis. This has led to numerous calculations of flux rates of ammonia (Aneja et al. 2001; Blunden and Aneja 2008). Emission rates have been calculated using a number of different methods such as enclosed flux chambers (e.g., Aneja et al. 2001; Blunden and Aneja 2008) and various micrometeorological techniques (e.g., Harper and Sharpe 2000; Phillips et al. 2004). Regardless of the method employed, the objective has been to incorporate variables such as pH, wind speed, and temperature into models so that ammonia emissions may be reasonably modeled under varied conditions.

Some of the odorants most strongly characteristic of animal wastes, however, simple phenols and indoles (Spoelstra 1977; Schiffman et al. 2001; Zahn et al. 2001), occur in relatively low levels and do not possess easily ionized functional groups that facilitate their capture. While ammonia may occur in lagoon wastes in concentrations over 1 g/L, malodorous aromatic compounds more typically occur in the lagoons in nanogram to microgram per liter levels (Merrill and Halverson 2002; Loughrin et al. 2008). Nevertheless, estimates of emission rates of these compounds from waste treatment structures are desirable so that the factors that control their release may be better understood.

In order to gain an appreciation of those factors that influence the release of two of these typical malodorants, *p*-cresol and *p*-ethylphenol, we entrained these compounds on thermal desorption tubes at two heights above a swine waste lagoon with simultaneous collection of meteorological data. These meteorological measurements were used to calculate heat fluxes and net radiation from the lagoon and the values analyzed for correlations with malodorous compound concentrations and gradients.

2 Experimental Methods

2.1 Site Description

Measurements were performed on an approximately 60 m^2 lagoon that served as the primary waste recipient for a farrowing operation of approximately 2,000 sows. Once a week, wastes were flushed from subfloor pits into the lagoon and fresh water was used to partially recharge the pits. There was no pit recharge from the lagoon. Typically, the lagoon was partially pumped down twice yearly, in the fall and spring, for application to crops. In the year of the study, the level of the lagoon was lowered by 10–20% in April. Agitation was used during pump down to remove as much as of the sludge layer as possible. Volatile collections were performed 11 times from 6 February to 30 March, 2009 and again eight times from 3 June to 25 June, 2009. Lagoon depth was about 2.9 m during the first collection period and 3 m during June.

2.2 Meteorological Measurements

Measurement of humidity, wind speed, and temperature at 0.5 and 1.5 m above the lagoon surface were taken by probes mounted on a float constructed from polyvinyl-chloride (PVC) pipe. The anemometers had an accuracy of 0.1 ms^{-1} in the range of 5–25 ms^{-1} and starting speed of 0.32 ms^{-1} while the temperature/humidity sensor had a range of 3–100% non-condensing for humidity with an accuracy of ±2% and a hysteresis of ±1% and a temperature range of −17 to 70°C with an accuracy of +0.5°C. The float was anchored to the lagoon by ropes tied to stakes at the side of the lagoon while a data cable led to a solar-powered data logger at the side of the lagoon. The data logger and sensors were all purchased from APRS World, LLC (Winona, MN, USA) and data were recorded in 5-min intervals.

Water temperatures were recorded at the lagoon surface, 0.3, 0.6, and 0.9 m deep, as well as in the sludge layer by HOBO® Pro V2 temperature sensors (Onset Computer Corp., Bourne, MA, USA) attached to a PVC pipe float. The sensors had a measurement range of −40°C to 70°C with an accuracy of ±0.18°C at 25°C and a resolution of 0.02°C. Data were recorded in 5-min intervals.

Meteorological measurements were also recorded 10 m to the south-west of the lagoon using a 15-channel HOBO weather station. Wind speed, direction, and gust speed were measured by a sensor mounted at a height of 3 m above the ground. Humidity and air temperature probes were placed in a solar radiation shield at a height of 2 m, and rainfall was measured with a tipping-bucket rain gage mounted at 3 m. Solar radiation at 2 m high

was measured with a silicon pyranometer with a spectral range of 300–1,100 m with a resolution 1.25 Wm^{-2} and an accuracy of 5%. The anemometer had a starting speed of 0.5 ms^{-1} with an accuracy of 0.5 ms^{-1} in the range of 17–30 ms^{-1} while the temperature/humidity sensor had a temperature range of −10 to 75°C with an accuracy of 0.2°C and a humidity range of 0–100% non-condensing with an accuracy of ±2.5% in the range of 10–90% relative humidity with a hysteresis of ±1%. Again, data was recorded in 5-min intervals.

2.3 Collection of Malodorous Compounds

Malodors were collected on thermal desorption tubes containing a 5 cm long bed of Tenax TA and 2 cm of Carboxen 569 (Supelco Inc., Bellefonte, PA, USA). Three tubes each were mounted at heights of 0.5 and 1.5 m on a float constructed of PVC pipe and was connected by tubing to 150 cm tall flow meters (Bel-Art Products, Pequannock, NJ, USA). The flow meters were connected via a manifold to a 12 VDC diaphragm pump (Gast Manufacturing, Inc., Benton Harbor, MI, USA) and 12 VDC portable power supply (W.W. Grainger, Inc., Chicago, IL, USA). Collections were of 2 h durations, and during sampling, flow rates were maintained at 250 $mL min^{-1}$. At the end of sampling, the tubes were placed in air-tight containers and kept cool until analyzed as described below.

2.4 Gas Chromatography–Mass Spectroscopy

Gas chromatography–mass spectroscopy was performed on a Gerstel model TDSA thermal desorption system and CIS 3 cooled injector (Gerstel USA Inc., Baltimore, MD, USA) interfaced to a Varian model 3800 gas chromatograph and Saturn 2200 ion trap (Varian Associates, Palo Alto, CA, USA). In the case of air samples, the thermal desorption tubes were desorbed in splitless mode using an initial temperature of 25°C with a delay time of 0.25 min and then heated at 60°C min^{-1} to 225°C with a final time of 3 min. Desorbed volatiles were transferred by a heated transfer line maintained at 240°C to a injection liner maintained at −50°C with liquid CO_2. After 5 min, compounds were transferred to a 30 m by 0.25 mm VF-23MS column (50% cyanopropylmethylpolysiloxane) with a film thickness of 0.25 μm (Varian Inc.) using a 20:1 split and heating the injector at 10°C s^{-1} to 300°C, a temperature that was held for 3 min. Further GC operating conditions were: column oven temperature maintained at 40°C for 2 min and then programmed at 2°C min^{-1} to 115°C and then at 15°C min^{-1} to 250°C and held for 10 min.

2.5 Wastewater Quality Analysis

Wastewater samples were collected using a variation of a technique described previously (Loughrin et al. 2010). Briefly, one pint (473 mL) glass jars were fitted with lids equipped with 2 two-way ball valves (United States Plastic Corp., Lima, OH, USA). Teflon tape was used on the jar threads to ensure a tight seal and one of the ball valves was connected to a check valve (Smart Products Inc., Morgan Hill, CA, USA). Peristaltic tubing, suspended in the lagoon at depths of 0.3 and 0.9 m, lead from the jars to a portable peristaltic pump (Cole-Palmer, Vernon Hills, IL, USA). Wastewater samples were taken at three locations within the lagoon for each depth.

Wastewater quality and dissolved/suspended gasses were measured as described (Loughrin et al. 2010).

2.6 Energy Flux Calculations

In order to understand atmospheric controls over the rate of lagoon emissions, latent heat fluxes were computed using an energy budget method and Bowen ratio estimates (e.g., Ham 1999; Brutsaert 2005; Quintanar et al. 2009). The technique is based on an estimate of the Bowen ratio from measurements of temperature and relative humidity at 0.5 and 1.5 m above the lagoon surface and at the lagoon surface. Relative humidity was assumed to be 100% at the lagoon surface. The technique also assumes that the eddy diffusivities for humidity and heat fluxes are the same. Given those measurements of relative and temperature, the Bowen ratio (B) was computed as:

$$B = \gamma\left(\frac{T(z1) - T(z2)}{e(z1) - e(z2)}\right) \quad (1)$$

Where e (from relative humidity measurements) and T are vapor pressure estimates and temperature measurements, respectively, taken at two heights (z_1 and z_2) above the lagoon surface and at the lagoon surface. The psychrometric constant (γ) is defined as

$\gamma = C_p p/(0.622\ L_e)$, where C_p is air heat capacity at constant pressure, p is atmospheric pressure and L_e is the latent heat of vaporization. In this study, emphasis was placed on evaporation which was directly computed from latent heat fluxes. This was estimated as:

$$E = \left(Q_{\mathrm{ne}} L_e^{-1}\right)(1+B)^{-1} \tag{2}$$

where Q_n is the net energy available at the lagoon surface and is defined as:

$$Q_n = R_n - G \tag{3}$$

with R_n being net solar radiation and G the lagoon heating rate estimated from the measured temperature profile of the water column.

In the absence of direct measurements we followed Brutsaert (2005) to estimate net radiation. To that end, the net radiation further decomposed as:

$$R_n = R_s(1-\alpha_{\mathrm{s}}) + \varepsilon_{\mathrm{s}} R_{\mathrm{ld}} - R_{\mathrm{lu}} \tag{4}$$

where R_s is the global short-wave radiation, α_{s} is the albedo of the water surface, R_{ld} is the incoming long-wave radiation, ε_{s} is the emissivity of the lagoon surface and R_{lu} is the outgoing long-wave radiation from the surface. The latter was estimated as:

$$R\mathrm{lu} = \varepsilon s \sigma T_s^4 \tag{5}$$

where σ is the Boltzmann constant and T_s is the absolute temperature of the lagoon surface. The R_{ld} could be expressed as:

$$Rld = \varepsilon ac \sigma T_a^4 \tag{6}$$

where $\varepsilon_{\mathrm{ac}}$ is the atmospheric emissivity and T_a was taken as the atmospheric temperature at 0.5 m. The $\varepsilon_{\mathrm{ac}}$ was represented as:

$$\varepsilon_{\mathrm{ac}=} A\left(e(z_2)/T_a\right)^B \tag{7}$$

with *e(z2)* the estimated vapor pressure at height z_2 (1.5 m), B=1/7, A=1.16 for the warm season measurements and A=1.28 for the cool season measurements. These values of A provided us with the estimates of atmospheric emissivity and subsequently R_n that provided us with good projections of malodorous compound concentration. It needs to be noted that Brutsaert (2005) prescribed A=1.31 for entire year and did not provide any seasonal values. However, Crawford and Duchon (1999) provided a methodology for estimating A that accounts for seasonal variations. We have also used the Crawford and Duchon method (Eq. 20) to calculate A and our estimates of seasonal A were comparable (Crawford and Duchon 1999). Side by-side comparison of Brutsaert with Crawford and Duchon provided superior correlations of heat fluxes with malodorous compound concentrations when we used the latter's method.

Data was analyzed in the SAS System for Windows v. 9.1 (SAS Institute Inc., Cary NC, USA 1996).

3 Results and Discussion

3.1 Meteorological Conditions and Wastewater Characteristics

During the cool season volatile collection period, weather conditions were quite variable as would be expected in February and March given Kentucky's humid subtropical climate (Fig. 1). Winds, for instance, were changeable and predominantly from the south with calms occurring only 2.25% of the time.

Temperatures and humidity were also variable (Table 1). Air temperature ranged from −3.85 to 22.1°C, while lagoon surface and sludge temperatures averaged 14.4°C and 11.0°C, respectively. Average humidity was low and ranged from 6.34 to 9.59 gm^{-3}. Insolation during the cool season volatile collections averaged 591 Wm^{-2}.

During the warm season collections, winds were calmer and predominately from the southwest (Fig. 1). In the case of both cool and warm season collections, wind readings from the swine waste lagoon weather station were in reasonable agreement with those obtained from a nearby Kentucky Mesonet weather station (Logan County, Kentucky Mesonet, 36.854 N, 86.92 W, http://www.kymesonet.org).

In the warm season, air, lagoon surface and sludge temperature averaged 28.3°C, 29.7°C, and 27.8°C, respectively (Table 1). As expected, humidity was much higher in the warm season, averaging 23.8 gm^{-3}. In the case of both cool and warm season collection campaigns, no condensing conditions occurred, averaging 55.7% and 66.7% relative humidity during the cool and warm seasons, respectively. During one warm season collection period, however, relative humidity reached as high as 97.8% (corresponding to 27.4 gm^{-3} water vapor). At no time was any condensation observed in the Tenax tubes, however. Average

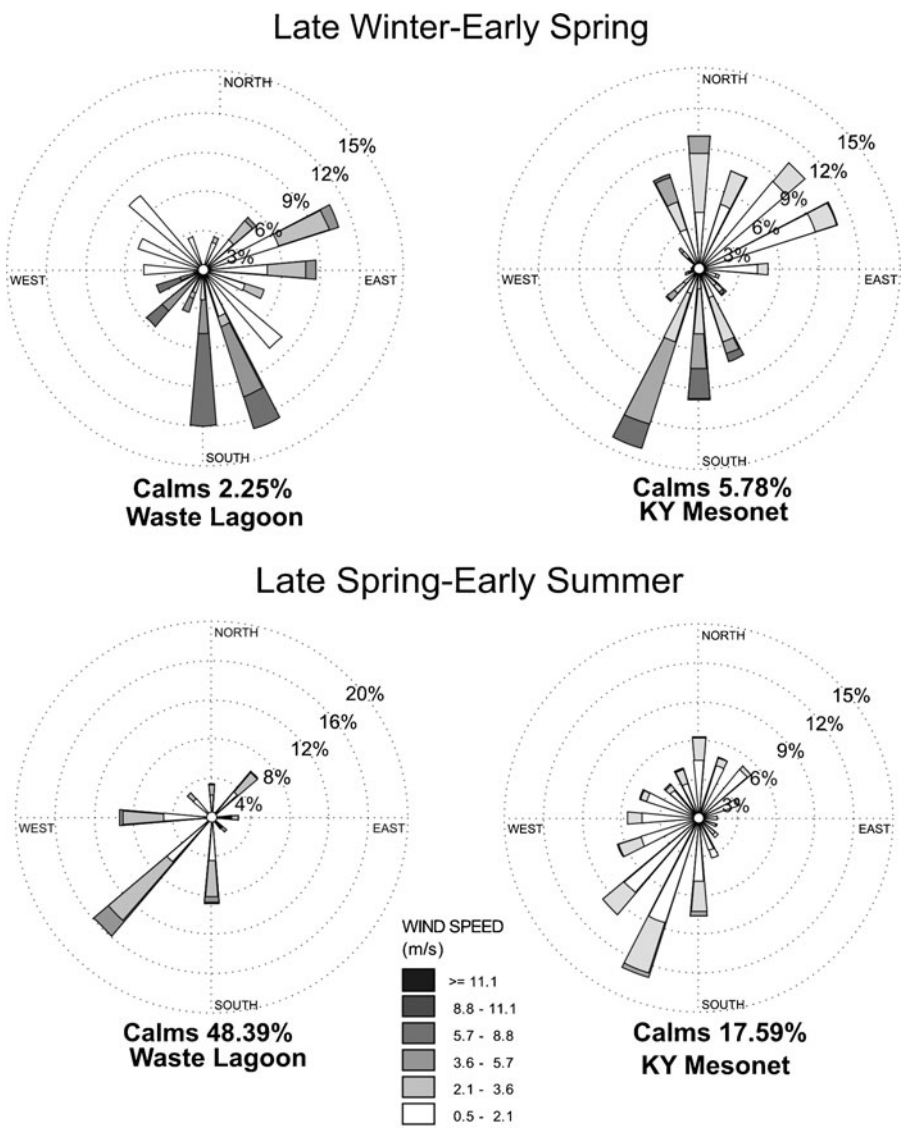

Fig. 1 Wind rose plots for wind speed and direction during volatile collections

insolation was slightly less than during the cool season collections at 572 Wm^{-2}.

Although there were marked differences in wastewater quality between the cool and warm season collection campaigns, these were not in general as pronounced as was the case in meteorological conditions (Fig. 2). Chemical oxygen demand and ammonia declined by approximately 62% and 35%, respectively, while organic carbon and nitrogen content declined by about 18% and 11%. Total suspended solids, on the other hand, increased by about 11% in the warm season. pH averaged 7.65 in the cool season and 7.87 in the warm season.

The wastewater concentrations of malodorants were dramatically lower during the warm season than they were during the cool season, however. *Para*-cresol and *p*-ethylphenol averaged 29,600 and 3,700 μgL^{-1}, respectively, in the cooler months and 1,460 and 331 μgL^{-1} in the warm season. Thus, there was markedly less malodorants available for volatilization during the warmer season. For instance, air concentrations of *p*-cresol at 0.5 m above the lagoon surface averaged 26,100 and 1,130 ngm^{-3} during the cool season and warm season collections, respectively. That declines in the concentrations of malodorous compounds in waste lagoons occur during the warm season is well known, and are most commonly thought to be due to a combination of factors including microbial catabolism (Gu and Berry 1991; Do et al. 2003)

Table 1 Descriptive statistics (mean and range) of meteorological conditions during volatile collections

Factor Meteorological conditions	Winter–early spring	Late spring–summer
Air temperature, (°C)	12.4 (−3.9 to 22.1)	28.3 (24.4 to 31.9)
Lagoon surface temperature, (°C)	14.4 (3.8 to 31.6)	29.7 (27.3 to 35.7)
Sludge temperature, (°C)	11.0 (3.7 to 15.7)	27.8 (25.1 to 30.9)
Insolation (Wm^{-2})	591.0 (148.1 to 964.4)	572.0 (75.6 to 1066.4)
Humidity (gm^{-3})	6.34 (1.53 to 9.59)	23.8 (9.0 to 26.7)
Evaporation ($mmm^{-2}h^{-1}$)	0.33 (−0.24 to 1.89)	5.28 (−0.51 to 8.48)
Net Radiation ($Wm^{-2}h^{-1}$)	173 (−111 to 677)	141 (−17.5 to 237)

and losses due to evaporation as the lagoons warm during the spring (Pfast and Fulhage 2000).

3.2 Lagoon Energy Budget and Malodorant Emissions

The model of choice was based on obtaining the largest correlation, as determined using PROC STEPWISE in SAS, between the concentration of the malodorants at 0.5 m above the lagoon surface as well as the gradient in their concentrations between 0.5 m and 1.5 m and derived meteorological variables such as latent and sensible heat fluxes, lagoon heating (G) and net radiation (R_n). The model that was most successful in explaining the observed variations in absolute concentrations and their vertical gradients was the model that included net radiation and evaporation jointly (Table 2).

While correlations with evaporation rates alone were rather high, it was found that this term on its own did not adequately explain variations in malodor concentrations and gradients. Thus, the R_n term was required. The coefficients for the regressions were:

$$p-\text{cresol}: \ \text{Concentration}_{0.5m} = -350 + (-3,080 \times \text{evaporation}) + (60.6 \times R_n) \quad (8)$$

$$\Delta\text{Concentration}_{0.5m-1.5m} = -1,890 + (-1,904 \times \text{evaporation}) + (40.9 \times R_n) \quad (9)$$

$$p-\text{ethylphenol}: \text{Concentration}_{0.5m} = 44.7 + (-376 \times \text{evaporation}) + (7.44 \times R_n) \quad (10)$$

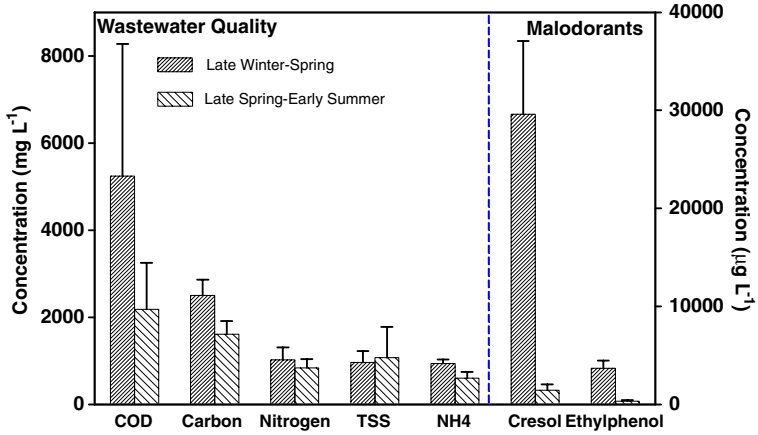

Fig. 2 Lagoon wastewater characteristics during late winter–early spring and late spring–early summer odor collection campaigns

Table 2 Results of statistical analyses for the determination of predictor variables for malodorant concentrations and gradients above the lagoon surface

		Compound Concentration at 0.5 m height		Δ Concentration 0.5–1. 5 m	
	Source	Pr>F	r^2	Pr>F	r^2
p-Cresol	Model	<0.0001	0.7369	<0.0001	0.7538
	Evaporation	0.0007	0.4979	0.0012	0.4696
	R_n	0.0015	0.2390	0.0006	0.2842
p-Ethylphenol	Model	<0.0001	0.7184	0.0002	0.6606
	Evaporation	0.0010	0.4834	0.0032	0.4081
	R_n	0.0021	0.2350	0.0033	0.2525

$$\Delta\text{Concentration}_{0.5m-1.5m} = -238 + (-239 \times \text{evaporation}) + (5.18 \times R_n) \quad (11)$$

where $\Delta\text{Concentration}_{0.5m-1.5m}$ is the difference in malodorant concentration at 0.5 and 1.5 m above the lagoon surface.

These regressions explained about 74% of the variations in *p*-cresol concentration and 72% of the variations in *p*-ethylphenol concentration above the lagoon surface, respectively, and were similarly successful in describing the variations in the malodor concentration gradients. As noted, the concentrations of *p*-cresol and *p*-ethylphenol in the lagoon wastewater were much lower in the warm season than during the cool season and in consequence the concentrations of malodorants in the air above the lagoon were also much lower (Fig. 3). Therefore, since the model included both cool season collections when evaporation rates on average were low, and warm season collections, when evaporation rates were relatively high, the evaporation terms have negative coefficients. It must be noted, nevertheless, when the cool season and warm season collection campaigns were

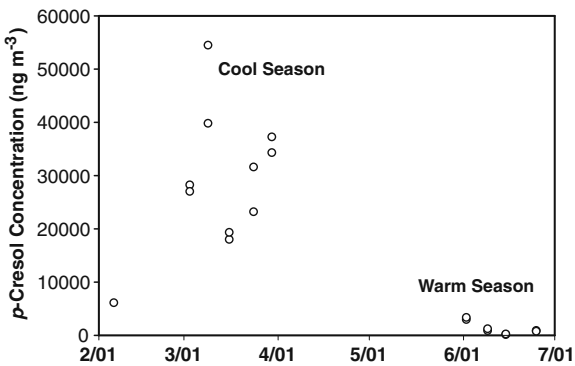

Fig. 3 *Para*-Cresol concentrations 0.5 m above the lagoon surface

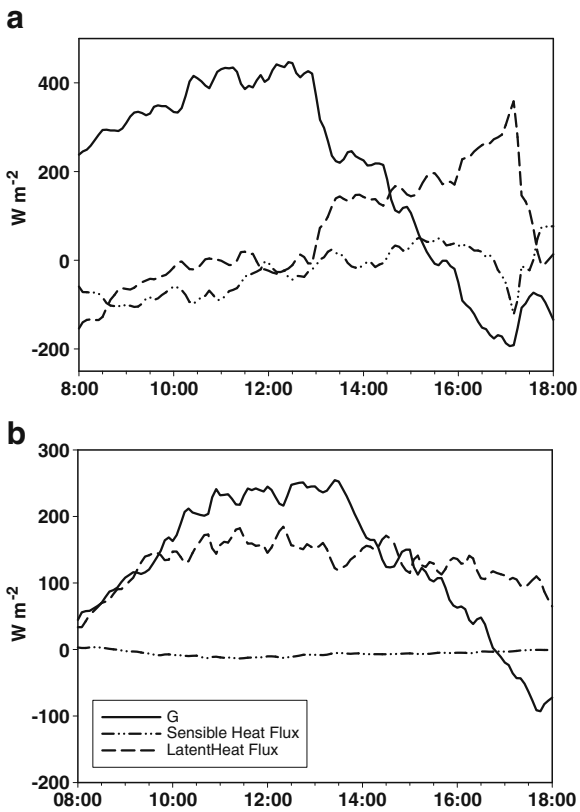

Fig. 4 Average daytime lagoon heating, latent and sensible heat fluxes during days on which samples were taken in the cool season (**a**) and warm season (**b**) odor collection campaigns

considered separately, the relationship of both evaporation and R_n to the variations in malodorant concentrations and gradients were positive.

As exemplified by the case of *p*-cresol, malodorant concentrations were much more sensitive to changes in net available lagoon energy than to changes in evaporation rates. As noted, the relationship between evaporation and malodorant concentrations was negative. Changes in evaporation rates did not affect malodorant emissions as profoundly as did changes in R_n, however. For instance, when the evaporation rate was held equal to zero, changes in R_n resulted in *p*-cresol concentrations at 0.5 m above the lagoon surface from 0 to over 30,000 $ng\,m^{-3}$. When the evaporation rate was held constant at 6 $mm\,h^{-1}$, *p*-cresol concentrations at that same height ranged from approximately 2,000 to over 15,000 $ng\,m^{-3}$.

While lagoon heating rates during midday, when the majority of volatile collections were done, were comparable during the cool season and warm season collection campaigns (Fig. 4), latent heat fluxes were much lower during the cool season. In fact, appreciable latent heat fluxes were not noted during days upon which volatile collections were performed until late in the afternoon. Given our findings, the greatest emission of malodorants from the lagoon should occur during this period during the cool season whereas emissions throughout the day are likely to be more constant during the warm season. It should also be noted that appreciable sensible heat fluxes were not detected during either season. This behavior has also been reported in studies in which water is not limited such as lakes and shallow lagoons (Rodríguez-Rodríguez and Moreno-Ostos 2006; Tanny et al. 2008).

3.3 Discussion of Findings and Conclusion

In conclusion, by the collection and measurement of malodorous compounds at two heights above the surface of a swine waste lagoon along with concomitant measurement of environmental parameters such as wind speed and temperature at the same heights, we were able to investigate the relationship of the emission of these compounds to the energy budget of the lagoon. We found that the emission *p*-cresol and *p*-ethylphenol from a swine waste lagoon are dependent on evaporation and the net available energy at the lagoon surface.

The results from heat flux calculations have been used in numerous instances to estimate fluxes of volatile compounds from numerous sources under varied situations. Just a few examples include ammonia emissions from waste lagoons (Phillips et al. 2004), terpenes, and other volatile emissions from forests (Ieda et al. 2006) and the volatilization of pesticides from bare fields and crops (Parmele et al. 1972). Results of the present work are being used as a means of selecting models so as to be able to estimate fluxes of malodorants from waste lagoons and thereby develop reasoned approaches to the mitigation of these nuisance compounds. For instance, shelterbelts of trees or shrubs have been proposed as a means of reducing emissions from animal rearing facilities (Tyndall and Grala 2006, 2009). While these shelterbelts are normally seen as a means of controlling emissions by reducing wind speeds, our results indicate that in the case of waste lagoons at least, shelterbelts might be more effective if placed so as to reduce exposure to the sun and hence lagoon heating.

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