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Gaseous Nitrate Radical: Possible Nighttime Atmospheric Sink for Biogenic Organic Compounds

Abstract. *The gaseous nitrate (NO_3) radical, which has recently been measured in nighttime ambient atmospheres over the United States and Europe at concentrations up to ~ 350 parts per trillion, has now been shown to react rapidly with the biogenically emitted organic compounds dimethyl sulfide (DMS), isoprene, and several monoterpenes. Computer simulations demonstrate that these reactions can dominate the atmospheric behavior of these organic compounds at night. Thus reaction with NO_3 radicals may be the unknown, nonphotochemical removal process for DMS recently invoked by Andreae and Raemdonck to explain the absence of a diurnal profile for DMS in maritime air influenced by continental air masses. Similarly, the nighttime reaction of NO_3 radicals with monoterpenes can be a dominant removal process, leading to very low monoterpene concentrations in ambient atmospheres during the early morning.*

The role of biogenically emitted organic compounds in the chemistry of both the polluted and the clean troposphere has received much attention (*1–5*). Sandberg *et al.* (*6*) and others (*7*) have proposed that hydrocarbons such as isoprene and the monoterpenes emitted from vegetation contribute significantly to photochemical air pollution, while dimethyl sulfide (DMS), emitted from oceans and other natural sources, is involved in the global atmospheric sulfur cycle (*8*), ultimately contributing to background acid deposition.

Current models of the atmospheric chemistry of organic compounds emitted from biogenic sources (*2, 9*) treat the consumption of such species as due solely to reaction with hydroxyl (OH) radicals or ozone (O_3) or to photolysis. However, serious discrepancies have been reported between the measured rates of emission from vegetation of isoprene and the monoterpenes and their observed ambient concentrations (*3, 4*). Moreover, Andreae and Raemdonck (*10*) have recently invoked an unknown, nonphotochemical removal process for DMS from "continentally influenced" marine air masses.

Within the past 3 years we and our collaborators (*11–14*) and Noxon and coworkers (*15*) have identified and measured the nitrate radical (NO_3) at night in both the clean and the polluted troposphere, using long pathlength (~ 1 to 17 km) differential optical absorption spectroscopy (DOAS). Recently, we have also measured the rate constants for the reaction in air of NO_3 radicals with a large number of organics (*16–18*) including DMS, isoprene, and selected monoterpenes. We report here calculations utilizing these kinetic and ambient concentration data, which show that nighttime reaction with the NO_3 radical is a dominant, previously unrecognized, atmospheric pathway for the removal of many organic compounds of biogenic origin.

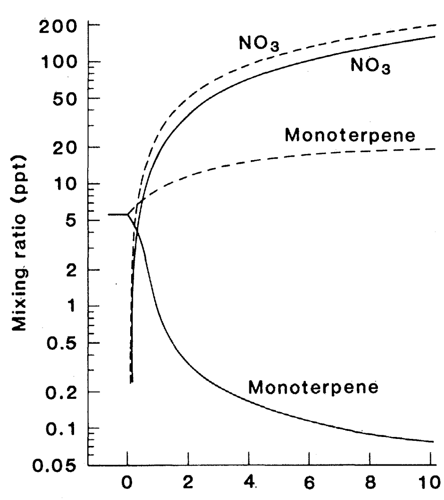


Fig. 1. Calculated time-concentration profiles for NO_3 radicals and monoterpenes for the Death Valley scenario (*21–24*). Solid lines are predictions for reaction between NO_3 radicals and monoterpenes; dashed lines are predictions for the assumption that there is no reaction.

Nighttime concentrations of NO_3 radicals measured in the United States (*11, 14*) and Europe (*13*) have ranged between the detection limit of the DOAS technique [~ 1 part per trillion (ppt)] and ~ 350 ppt, with typical concentrations in continental air masses ranging between ~ 10 and ~ 100 ppt. In polluted atmospheres the NO_3 radical concentrations increase (*11*) after sunset to a peak at ~ 2000 hours and then decrease rapidly to below the detection limit by about midnight. In contrast, in semiarid desert atmospheres the NO_3 radical concentrations generally increase (*14*) after sunset to a plateau value which persists until sunrise; after sunrise, the concentrations decline rapidly to less than 1 ppt because of the large photolytic cross section (*19*) of the NO_3 radical.

The rate constants (*20*) we have recently determined at room temperature for the gas-phase reactions of NO_3 radicals with biogenic organics are fast, ranging from ~ 5×10^{-13} cm^3 sec^{-1} per molecule for DMS (*17*) and isoprene (*18*) to (1 to 8) $\times 10^{-12}$ cm^3 sec^{-1} per molecule for the monoterpenes α- and β-pinene, *d*-limonene, and Δ^3-carene (*18*). To assess the importance of these reactions relative to reaction with OH radicals or with O_3, we have calculated the atmospheric lifetimes, τ, of DMS, isoprene, and selected monoterpenes for these three reaction pathways under two sets of atmospheric conditions (Table 1).

Reaction with NO_3 radicals is the dominant loss process for DMS and the monoterpenes, even under conditions corresponding to the clean troposphere (Table 1); for isoprene, consumption by NO_3 radicals at night is equal in importance to loss due to attack by OH radicals during daylight hours. In moderately polluted atmospheres, the reaction of NO_3 radicals with the monoterpenes leads to extremely short monoterpene lifetimes of ~ 1 to 5 minutes (Table 1). Indeed, the rate of *d*-limonene consumption by NO_3 radicals at night is about ten times that of reaction with O_3 and ~ 30 times that of daytime reaction with OH radicals. Clearly, reactions of NO_3 radicals with DMS, isoprene, and the monoterpenes significantly reduce the nighttime ambient concentrations of these organics relative to those that would occur if the NO_3 radical reactions are not considered, as is the case for current chemical models of the clean and polluted troposphere (*2, 3, 8, 9*).

In order to quantitatively illustrate the magnitude of the effects of these reactions, we have calculated NO_3 radical and monoterpene concentrations during nighttime hours, utilizing an appropriate chemical mechanism (*21, 22*). Calculations were carried out for conditions representative of the locations for which we have spectroscopically measured ambient NO_3 radical concentrations [Death Valley, Whitewater, Phelan, and Edwards Air Force Base, California (*14*), and Claremont and Riverside, California (*11*)]. The NO_2, O_3, and H_2O concentrations and the temperatures and relative humidities representative of these locations were obtained from ambient data (*11, 14, 23*). Monoterpene emission rates (*24*) were derived from the emission rate data of Zimmerman (*25*) and Winer *et al.* (*5*). A box model was used, with a constant inversion height of 1000 m and with constant NO_2, O_3, and H_2O concentrations and monoterpene emission rates.

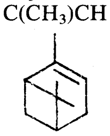
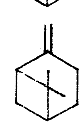
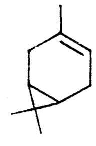
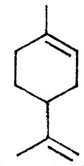
For simulations in which reaction between NO_3 radicals and the monoterpenes was permitted, the calculated time-concentration profiles of the NO_3 radical exhibited a rapid rise after sunset to a plateau value which persisted until sunrise (Fig. 1). This behavior was in good agreement with our spectroscopic observations at Whitewater, Phelan, Edwards Air Force Base, and Death Valley (*14*), and the plateau NO_3 radical concentration shown in Fig. 1 lies within the range observed at these semiarid desert sites (*14*). The calculated monoterpene concentrations declined rapidly after sunset (Fig. 1) to values in the range 0.08 to 1.2 ppt (depending upon location) just prior to sunrise.

When no reaction between NO_3 radicals and the monoterpenes was permitted, the calculated time-concentration profiles of the NO_3 radical remained little changed from the previous case. However, the calculated monoterpene concentrations rose monotonically from their sunset values to ~ 20 to 150 ppt just prior to sunrise. Thus, inclusion of the reactions of NO_3 radicals with the monoterpenes leads to a dramatic decrease of two to three orders of magnitude in the predicted ambient concentrations of the monoterpenes at night under these atmospheric conditions (clean to moderately polluted) but has little effect on the NO_3 radical profiles.

In order to generalize these simulations to a wide range of atmospheric conditions, we carried out further calculations (*26*) in which the NO_2 concentrations varied from 0.05 to 25 parts per billion (ppb), while the monoterpene emission rates varied over three orders of magnitude to encompass values reported or expected for vegetation densities ranging from desert to forested areas (from 15 to ~ 7700 $\mu g\ m^{-2}\ hour^{-1}$).

The results of these calculations are shown in Fig. 2. The results for Death Valley and the other locations discussed earlier fall on the plateau region where the formation rate of NO_3 radicals exceeds the monoterpene emission rate. Under these conditions the reaction of NO_3 radicals with monoterpenes will be a dominant loss process for the monoterpenes at night, leading to a large reduction in monoterpene concentrations just prior to sunrise [by a factor of ~ 100 to 150 for the conditions of O_3 and H_2O vapor concentrations and temperature selected (*26*)]. In these calculations we did not take into account the possible presence of other organics which react rapidly with the NO_3 radical [for example, hydroxy-substituted aromatics and the tri- and tetrasubstituted alkenes (*16*)]. Thus for urban atmospheres that contain such organics the monoterpene reductions shown in the corresponding portion (*23, 24*) of Fig. 2 would represent upper limits to the actual case (although no data are available to show that such reactive organics of anthropogenic origin are present during nighttime hours at concentrations leading to any significant scavenging of NO_3 radicals).

Table 1. Calculated lifetimes of dimethyl sulfide, isoprene, and four monoterpenes due to reaction with O_3 or OH radicals or NO_3 radicals in "clean" and "moderately" polluted atmospheres.

Organic	Structure	Organic lifetime*					
		"Clean" atmosphere†			"Moderately polluted" atmosphere‡		
		τ_{O_3}, 24 hours	τ_{OH}, daytime (hours)	τ_{NO_3}, nighttime (min)	τ_{O_3}, 24 hours	τ_{OH}, daytime (min)	τ_{NO_3}, nighttime (min)
Dimethyl sulfide	CH_3SCH_3	> 20 days	28	120	> 3 days	420	13
Isoprene	$CH_2=C(CH_3)CH=CH_2$	32 hours	2.9	216	10 hours	44	22
α-Pinene		4.6 hours	4.6	20	1.4 hours	72	2
β-Pinene		18 hours	3.6	50	5.5 hours	54	5
Δ^3-Carene		3.2 hours	3.5	12	1.0 hour	53	1.2
d-Limonene		36 minutes	2.0	9	11 minutes	30	0.9

*Organic lifetime, τ, is given by $\tau_x = 1/k_x[X]$, where k_x and $[X]$ are the reaction rate constants and ambient concentrations, respectively, of OH radicals or NO_3 radicals or O_3. Rate constants used are from the following: for NO_3 radicals (*17, 18*); for O_3 (*22, 31*); for OH radicals (*17, 32*). †Assuming 30 ppb of O_3 (24-hour average), $1 \times 10^6\ cm^{-3}$ (0.04 ppt) average concentration of OH radicals during daylight hours, and 10 ppt of NO_3 during nighttime hours. ‡Assuming 200 ppb of O_3 (24-hour average), $4 \times 10^6\ cm^{-3}$ (0.16 ppt) average concentration of OH radicals during daylight hours, and 100 ppt of NO_3 during nighttime hours.

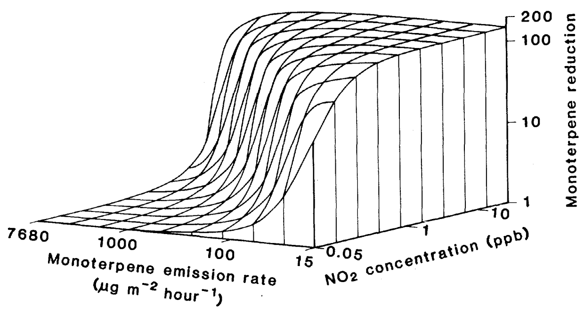


Fig. 2. Plot of the ratio (sunrise monoterpene concentration in the absence of reaction with NO_3 radicals)/(sunrise monoterpene concentration in the presence of reaction with NO_3 radicals) (labeled monoterpene reduction) as a function of NO_2 concentration and monoterpene emission rate. The NO_3 formation rate is directly proportional to the NO_2 concentration for a constant O_3 concentration.

The opposite case, monoterpene emission rates greater than the NO_3 radical formation rates, leads to the behavior shown in the lower left corner of Fig. 2. An example of this case, that is, of an essentially unchanged monoterpene time-concentration profile with and without NO_3 radical reactions, is a coniferous forest in an area free of atmospheric pollution. The transition between the two plateau regions in Fig. 2 occurs when the NO_3 radical formation rate is within a factor of 2 of the monoterpene emission rate and corresponds to the "cliff" in this figure. The results of these calculations clearly demonstrate that reactions of NO_3 radicals may explain, at least in part, the widely reported discrepancy between measured ambient monoterpene concentrations and those predicted on the basis of emission rates (*3–5*).

In the case of DMS, Andreae and Raemdonck (*10*) recently reported that over the remote equatorial Pacific Ocean DMS concentrations exhibited a diurnal profile. Concentrations ranged from ~ 50 to ~ 80 ppt (average, ~ 70 ppt), with the higher concentrations during nighttime hours. However, in atmospheres over the Atlantic and Pacific oceans and the Gulf of Mexico, which they reported to be influenced by continental air masses, much lower (2.5 to 17 ppt) concentrations of DMS were observed, with no diurnal variation. Andreae and Raemdonck suggested that "in continentally influenced air masses there is an additional removal process for DMS that is not directly photochemical" (*10*, p. 746).

We propose that this unknown loss process involves the nighttime reaction between the NO_3 radical and DMS. To test this, we carried out simulations corresponding to the two characteristic air masses found at the locations they studied (*10*). For the clean troposphere over the Pacific we chose NO_2 concentrations of the order of 10 to 30 ppt (*27, 28*) and ambient O_3 concentrations of ~ 30 ppb (*28*). These concentrations lead to a low NO_3 radical formation rate and hence a low atmospheric concentration. Under these conditions our calculations (*29*) predict that the reaction of the NO_3 radical with DMS will have only a small effect on the DMS nighttime concentration (Fig. 3) and that the maximum nighttime concentration of NO_3 radical will be ~ 0.3 ppt. Thus, for clean maritime air DMS will be essentially removed only by OH radicals during daylight hours. Indeed, our calculated diurnal range of DMS concentrations of ~ 60 to ~ 140 ppt is reasonably consistent with the profile reported by Andreae and Raemdonck for remote Pacific regions (*10*).

On the other hand, maritime atmospheres influenced by continental air masses may contain significantly higher concentrations of NO_2, which lead to correspondingly higher concentrations of NO_3 radicals at night. In this case the nighttime reaction of DMS with NO_3 radicals can become an important factor in removing DMS (*30*). Indeed, an NO_3 radical concentration of ~ 3 ppt would be as important a sink for DMS during the night (Fig. 3) as an OH radical concentration of 4×10^6 cm^{-3} during the day (a plausible average 12-hour value for the Pacific Ocean at equatorial latitudes). Nighttime concentrations of NO_3 radicals higher than ~ 3 ppt would lead to correspondingly lower nighttime DMS concentrations; conversely, lower NO_3 radical concentrations would permit accumulation of DMS during the night (Fig. 3). Thus only modest concentrations of NO_3 radicals, at the low end of the range observed to date in continental air masses, are required to account for the absence of a diurnal variation in DMS concentrations for offshore maritime atmospheres (*10*).

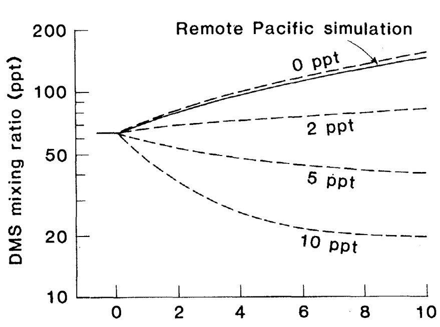


Fig. 3. Calculated time-concentration profiles for dimethyl sulfide for constant NO_3 radical concentrations of 0, 2, 5, and 10 ppt (dashed lines) and for a simulated (*29*) remote Pacific atmosphere (solid line).

Clearly, the NO_3 radical is a key atmospheric constituent and its nighttime reactions with certain important biogenically emitted organic compounds should be incorporated into chemical models of the troposphere.

ARTHUR M. WINER
ROGER ATKINSON
JAMES N. PITTS, JR.
Statewide Air Pollution Research Center, University of California, Riverside 92521

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$$NO + O_3 \rightarrow NO_2 + O_2 \quad (1)$$
$$NO_2 + O_3 \rightarrow NO_3 + O_2 \quad (2)$$
$$NO_2 + NO_3 \rightarrow N_2O_5 \quad (3)$$
$$N_2O_5 \rightarrow NO_2 + NO_3 \quad (4)$$
$$NO_2 + NO_3 \rightarrow NO + NO_2 + O_2 \quad (5)$$
$$NO + NO_3 \rightarrow 2\ NO_2 \quad (6)$$
$$N_2O_5 + H_2O \rightarrow 2\ HNO_3 \quad (7)$$
$$NO_3 + \text{terpene} \rightarrow \text{products} \quad (8)$$
$$O_3 + \text{terpene} \rightarrow \text{products} \quad (9)$$

Rate constants for reactions 1 through 6 were taken from recent evaluations [R. Atkinson and A. C. Lloyd, *J. Phys. Chem. Ref. Data*, in press; *Chemical Kinetics and Photochemical Data for Use in Stratospheric Modeling* (NASA Evaluation 5, Jet Propulsion Laboratory publication 82-57, Pasadena, 1982)]. The rate constant for reaction 7 of 1.3×10^{-21} cm^3 sec^{-1} per molecule was taken from E. C. Tuazon, R. Atkinson, C. N. Plum, A. M. Winer, J. N. Pitts, Jr., *Geophys. Res. Lett.* **10**, 953 (1983). A rate constant of 5×10^{-12} cm^3 sec^{-1} per molecule was chosen for reaction 8 (*18*). The rate constant for reaction 9 was taken to be 1×10^{-16} cm^3 sec^{-1} per molecule, independent of temperature over a small temperature range around 298 K, a value consistent with available kinetic data (*22*). Sunset was assumed to occur instantaneously, an approximation that had no detectable effect on the model predictions. We derived the initial monoterpene concentrations at sunset by solving the steady-state equation involving the monoterpene emission rates and the daylight monoterpene loss processes by reaction with OH radicals (assumed to be at a constant concentration of 2×10^6 cm^{-3}) and O_3.
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23. The NO_2, O_3, and H_2O concentrations and temperature, respectively, chosen for these locations were as follows: Death Valley, 0.3 ppb, 30 ppb, 4 torr, and 303 K; Whitewater and Phelan, 3 ppb, 30 ppb, 6 torr, and 290 K: Edwards Air Force Base, 1 ppb, 30 ppb, 9 torr, and 300 K; Riverside and Claremont, 40 ppb, 40 ppb, 15 torr, and 303 K.
24. A monoterpene emission rate of 300 μg m^{-2} $hour^{-1}$ for Riverside and Claremont was based on the upper limits for average nighttime monoterpene emissions determined for the Los Angeles area by Winer *et al.* (*5*). Monoterpene emission rates for Whitewater and Phelan of 150 μg m^{-2} $hour^{-1}$ were based on the value for nonconifer, nonisoprene species given for desert areas by Zimmerman (*25*). An emission rate of 30 μg m^{-2} $hour^{-1}$ for Death Valley and Edwards Air Force Base was arbitrarily chosen as a conservative upper limit for these largely nonvegetated areas.
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$$OH + DMS \rightarrow \text{products} \quad (10)$$

and

$$NO_3 + DMS \rightarrow \text{products} \quad (11)$$

with a rate constant of 1×10^{-11} cm^3 sec^{-1} per molecule for reaction 10 and a rate constant of 5.4×10^{-13} cm^3 sec^{-1} per molecule for reaction 11 (*17*).
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33. This work was supported in part by National Science Foundation Atmospheric Chemistry Branch grant ATM-8209028 and Environmental Protection Agency grant R-810917. We thank B. J. Finlayson-Pitts for helpful discussions and M. Poe for generating Fig. 2.

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Cytomegalovirus Replicates in Differentiated but Not in Undifferentiated Human Embryonal Carcinoma Cells

Abstract. *To study the mode of action of human cytomegalovirus, an important teratogenic agent in human populations, the susceptibility of a pluripotent human embryonal carcinoma cell line to the virus was investigated. Viral antigens were not expressed nor was infectious virus produced by human embryonal carcinoma cells after infection, although the virus was able to penetrate these cells. In contrast, retinoic acid–induced differentiated derivatives of embryonal carcinoma cells were permissive for antigen expression and infectious virus production. Replication of human cytomegalovirus in human teratocarcinoma cells may therefore depend on cellular functions associated with differentiation.*

Intrauterine infection by human cytomegalovirus (HCMV) is a common cause of abnormal embryogenesis and fetal death (*1*). In the United States HCMV has been implicated in 2700 to 7600 cases of congenital birth defects annually (*2*). The mechanism and timing of fetal infection are obscure, since maternal infections are not usually clinically apparent, but HCMV transmission and severe damage to the fetus may occur very early in pregnancy (*3*). Also, only human fibroblasts are normally susceptible to productive infection by HCMV in vitro, and little is known about the control of cellular susceptibility to HCMV infection, particularly in the human embryo. Since embryonal carcinoma (EC) cells, the stem cells of teratocarcinomas, are thought to resemble cells of the early embryo in their biochemical and developmental properties (*4*), the recent characterization of a pluripotent human EC cell line now permits experimental study of the growth of HCMV in a cell akin to early human embryonic cells.

Line NT2/B9, a clonal human EC cell line isolated from TERA-2 (*5*), exhibits a

Table 1. Susceptibility of NT2/B9 EC and RA-induced differentiated cells to HCMV infection, as determined from the expression of viral antigens and the production of infectious virus. Cells were grown, infected, and tested for the presence of viral antigens by immunofluorescence as described in the legend to Fig. 1. The cells (3×10^3 to 5×10^3) were counted in each preparation to determine the percentage of cells positive for viral antigen. The number of cells containing infectious virus was determined by an infectious center assay. Infected NT2/B9 cells were washed five times in phosphate-buffered saline (PBS), trypsinized, and counted. Cells (3×10^2) were incubated with an excess of antiserum to HCMV (1 ml; 1:8 dilution of the antiserum and 1:24 dilution of guinea pig complement) for 1 hour, washed three times with PBS, and seeded on three parallel cultures of confluent MRC-5 monolayers at 1×10^2 NT2/B9 cells per petri dish. Cultures were overlaid with agar and the plaques were counted after 2 weeks. The mean number of plaques in the three parallel cultures are given. The immune serum fully neutralized approximately 3000 infectious particles at a dilution of 1:32 in separate experiments. The yield of virus in the culture medium was determined by the method of Wentworth and French (*19*). Expression of SSEA-3 was detected by reactivity of the cells with a specific monoclonal antibody assayed by flow cytofluorometry (*5*). Abbreviation: PFU, plaque-forming units.

Cells	Cells containing viral antigen (percent)			Infectious virus 7 days after infection in		Cells expressing SSEA-3 at time of infection (percent)
	Days after infection			Intact cells (PFU per 10^2 cells)	Culture medium (PFU/ml)	
	2	4	7			
NT2/B9	1.2	1.4	1.8	0.03	12	94.5
NT2/B9 + RA	68	76	85	45	5×10^3	3.2