Table 5. Chemical NO_x Budget in the Free Troposphere: Observations and Models

Field Experiment	$NO + NO_2$	HNO_3	PAN	NO_x Budget	Notes	Reference
				Aircraft Studies		
ABLE 3A, July/Aug. 1988 50°-75°N, 150°-170°W, 5-6.2 km	20	100	320	$((P-L)_{HNO_3}/(L-P)_{PAN} = 5.2)$	1, 10, 11, 12	*Jacob et al.* [1992]
ABLE 3B, July/Aug. 1990 45°-57°N, 65°-90°W, 2.5-6.2 km	33	50	240	$(L_{NO_x}/P_{NO_x} = 0.95)$	2, 10, 11, 12	*Fan et al.* [1994]
TRACE-A, Oct. 1992 40°S-20°N, 60°W-40°E, 4-8 km	57	130	294	$(L_{NO_x}/P_{NO_x} = 3.6)$	3, 10, 11, 13	*Jacob et al.* [1996]
ibid., 8-12 km	145	59	223	$(L_{NO_x}/P_{NO_x} = 5.6)$	3, 10, 11, 13	*ibid.*
PEM-West A, Oct. 1991 0-25°N, 100°E-140°W, 7-13 km	∼27	12	16	$R_N < 0.7$ (3-5)	4, 13	*Singh et al.* [1996a]
PEM-West B, Feb/Mar. 1994 110°E-180°, 5°S-25°N, 7-12 km	84	54	39	$R_N = 0.64$ (∼2)	5, 10, 11, 13	*Kondo et al.* [1997]
PEM-West B, Feb/Mar. 1994 110°E-180°, 5°S-10°N, 6-12 km	70 (45)	160 (95)	25 (40)	R_N∼2.9 (2.8)	6, 12	*Singh et al.* [1998]
PEM-Tropics A, Sep/Oct. 1996 0-30°S, 160°E-90°W, 6-12 km	21	48	27	$(L_{NO_x}/P_{NO_x} = 1.9)$	10, 11, 13	*this study*
				Mountain Site Measurements		
MLOPEX I, spring 1988 18°N, 156°W, 3.4 km	32 (20)	116 (184)	17 (15)	$R_N = 3.5$ (∼17, 9.4)	7, 12	*Liu et al.* [1992], *Brasseur et al.* [1996]
MLOPEX IIa, fall 1991	26 (23)	76 (232)	12 (13)	$R_N = 2.0$ (10, 21)	8, 12	*Brasseur et al.* [1996], *Hauglustaine et al.* [1996]
MLOPEX IIb, winter 1992	33 (29)	78 (155)	29 (28)	$R_N = 1.4$ (5.9, 21)	8, 12	*ibid.*
MLOPEX IIc, spring 1992	41 (19)	140 (170)	36 (17)	$R_N = 3.2$ (8.8, 24)	8, 12	*ibid.*
MLOPEX IId, summer 1992	30 (25)	84 (351)	9 (11)	$R_N = 2.4$ (14, 24)	8, 12	*ibid.*
Tenerife, summer 1993 28°N, 16°W, 2.4 km	70 (49)	410 (380)	10 (14)	$R_N = 5.9$ (7.8)	9	*Schultz et al.* [1998]

Values in parantheses are model results. The table compiles mean or median concentrations of NO_x, HNO_3, and PAN observed in previous field studies of the remote troposphere, together with summary results of chemical NO_x budget analyses conducted using photochemical point models or in some cases global three-dimensional models. Some models use the $HNO_3/(NO + NO_2)$ concentration ratio (R_N) as diagnostic of the NO_x budget while others use the ratio of chemical loss of NO_x (L_{NO_x}) to chemical production (P_{NO_x}) when the model is constrained with local observations. Notes are as follows: (1) No heterogeneous NO_x loss. The ratio of net loss of PAN to net production of HNO_3 exaggerates the NO_x budget imbalance. (2) Value for background conditions. Concentrations are weighted averages over three altitude bands. L_{NO_x}/P_{NO_x} is read from figure and corresponds to mean(L_{NO_x})/mean(P_{NO_x}). (3) N_2O_5 hydrolysis in aerosols included. (4) The sum of mean NO and mean NO_2 given by *Singh et al.* [1996a] is ∼58 pptv, but *Crawford et al.* [1996] conclude that observed NO_2 is high by a factor of 3-4. The values in this table include mean NO_2 reduced by a factor of 3. Observed R_N taken from *Singh et al.* [1996b, Figure 5]. Modeled R_N from the MOGUNTIA global three-dimensional model which includes N_2O_5 hydrolysis in aerosols. (5) Entry for air masses that were classified as "maritime tropical". *Crawford et al.* [1997] state that recycling from HNO_3 contributes 32% of the required NO_x source for the complete upper tropospheric PEM West B data. (6) Average of observations and model results binned in 5° latitude intervals, taken from *Singh et al.* [1998, Figures 6 and 9]. Global three-dimensional model from *Wang et al.* [1998]. (7) Model concentrations from MOZART global three-dimensional model [*Brasseur et al.*, 1996]. First modeled R_N from *Liu et al.* [1992]: photochemical steady state box model with no heterogeneous NO_x loss. Second modeled R_N from the MOZART model including N_2O_5 hydrolysis in aerosols. In a third analysis of MLOPEX I data, *Chatfield* [1994] gives a range from 14-25 for simulated R_N from a Lagrangian box model with N_2O_5 hydrolysis. (8) Model concentrations and first modeled R_N values are from *Brasseur et al.* [1996] who report results from the MOZART global three-dimensional model which includes N_2O_5 hydrolysis. Second modeled R_N from *Hauglustaine et al.* [1996]: Lagrangian box model with heterogeneous N_2O_5 hydrolysis ($\gamma = 0.1$) and aerosol surface areas estimated from local aerosol scattering coefficient measurements. (9) HNO_3 estimated as $NO_y - NO_x -$ PAN. Case study with lagrangian box model along descending trajectory. No heterogeneous NO_x loss. (10) NO_2 from photochemical steady state. (11) Photochemical box model in diurnal steady state constrained by observed NO, HNO_3, and PAN. (12) Mean values. (13) Median values.