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Influence of vertical transport on free tropospheric aerosols over the central USA in springtime

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Abstract. Measurements of the atmospheric aerosol chemical composition during the Subsonic Aircraft: Contrail and Cloud Effects Special Study (SUCCESS) indicate substantial vertical transport of boundary layer aerosol to the free troposphere over the south-central United States during springtime. Mixing ratios of water-soluble aerosol Ca^{2+} at 6 -12 km altitude exhibited a median mixing ratio of 20 pptv, with 15% of the measurements ≥100 pptv and a maximum of 1235 pptv. In air parcels with enhanced Ca^{2+}, the ratios K^+/Ca^{2+}, Mg^{2+}/Ca^{2+}, and Na^+/Ca^{2+} in the bulk aerosol were distinctly characteristic of those in limestone and/or cement. Significantly enhanced mixing ratios of aerosol SO_4^{2-}, NO_3^-, and NH_4^+ were also concomitant with the elevated Ca^{2+}, suggesting transport of both crustal and anthropogenic aerosols to the upper troposphere. The mass concentration of water-soluble aerosol material was in the range 0.1 - 6 μg m^{-3} STP, and estimated crustal dust levels were 7 - 160 μg m^{-3} STP.

Introduction

Transport of boundary layer chemical constituents to the free troposphere occurs in association with deep convective systems. Over the central and southwestern United States during spring and summertime this process is important for determining the vertical distribution of many trace gases including O_3, CO, NO_x (NO + NO_2), and hydrocarbons [*Dickerson et al.*, 1987; *Luke et al.*, 1992; *Ridley et al.*, 1994; *Ridley et al.*, 1996]. Since the lifetime of NO_x in the upper troposphere is much longer than at lower altitudes, transport of reactive species by convective systems could have important implications for photochemical production of O_3 and partitioning of HO_x (HO + HO_2) and reactive nitrogen (NO + NO_2 + PAN +HNO_3 + ...) aloft.

Dust storms also occur in springtime over the central and southwestern United States. Indeed, wind-generated soil material is the dominant form of primary continental particles [*Gillette*, 1980]. The size distribution of soil-derived particles in the troposphere is bimodal, with large particles exhibiting a mode around 50 μm diameter and small ones in the 1 - 10 μm range [*Patterson and Gillette*, 1977]. Measurements of aerosol composition as a function of altitude over the southwestern United States indicate that calcium is a good tracer for soil material in the troposphere [*Gillette and Bilifford*, 1971]. We have adopted this same tracer convention here to facilitate the interpretation of aerosol data obtained on aircraft flights conducted in March-April 1996 over the central United States during the SUbsonic aircraft: Contrail and Cloud Effects Special Study (SUCCESS).

Methods

SUCCESS was conducted in March-April 1996 with the primary study area located over the Department of Energy's Clouds and Radiation Testbed (CART) site located in northern Oklahoma and southern Kansas. A series of 12 flights were focused on studying cirrus cloud formation and their radiative properties plus the potential influence on these processes by subsonic aircraft emissions. Three flights also took place over the eastern Pacific Ocean off the California coast. Our atmospheric aerosol sampling system was flown on the NASA Ames DC-8 research aircraft platform which was based in Salina, Kansas, during the experiment.

Aerosol samples were collected with 10-15 minute time resolution on Zefluor Teflon filters for determination of water-soluble ions (Cl^-, NO_3^-, SO_4^{2-}, $C_2O_4^{2-}$, Na^+, K^+, NH_4^+, Ca^{2+}, and Mg^{2+}) and on glass fiber filters for measurement of ^7Be and ^{210}Pb activites. We used a tandem inlet assembly consisting of curved leading nozzles housed in shrouds to provide isoaxial, isokinetic sampling [*Dibb et al.*, 1996].

The handling procedure for the Teflon filters involved: (1) packing the filter cassettes at the University of New Hampshire in micro-clean bags, (2) exposure of the filters for various sampling intervals, (3) purging of the clean bags with ultra zero air as the filters were sealed in them immediately after sampling, (4) immediate freezing of collected samples to -30° C, and (5) overnight shipment of frozen samples to the University of New Hampshire for chemical analysis within 48 hours of collection. Details of the sampling and analytical procedures have been described previously [*Talbot et al.*, 1992; *Dibb et al.*, 1996].

Results

The vertical distribution of aerosol Ca^{2+} is shown in Figure 1 for all 14 flights conducted over the central United States. In the 6 -12 km altitude range the median Ca^{2+} mixing ratio was 20 pptv, with a mean and one standard deviation of 63 ± 145 pptv (n = 200). There was a significant number of observations with Ca^{2+} enhanced over this median value; above 6 km altitude 15% of the data exhibited Ca^{2+} mixing ratios of ≥100 pptv. Mixing ratios of aerosol Ca^{2+} above ≈100 pptv were coincident with significantly enhanced levels of CO and NO. A median mixing ratio for Ca^{2+} of 20 pptv is identical to the values observed on SUCCESS flights flown over the eastern Pacific both before and after the primary flight series over the central United States. This value is also very similar to data we obtained over this same area of the eastern Pacific during the NASA Global Tropospheric Experiment PEM-West A & B flight series in 1991 and 1994 (median Ca^{2+} = 24 pptv). Together these data indicate that 20 pptv is a representative mixing ratio of Ca^{2+} in the westerly flow of aged air parcels entering North America from over the eastern Pacific.

The three largest mixing ratios of Ca^{2+} above 6 km altitude were observed in the 11-12 km range. One of these, 460 pptv Ca^{2+}, was measured in a region influenced by stratospheric inputs (11.8 km altitude) where the coincident concentration of ^7Be was 10,000 fCi m^{-3} STP. The vertical temperature profile as measured along the DC-8 flight track with the microwave temperature profiler indicated that we flew 1 - 1.5 km above the thermal tropopause. It is likely that the enhanced Ca^{2+} was due to vertical transport of boundary layer air to the upper troposphere. On numerous occasions during

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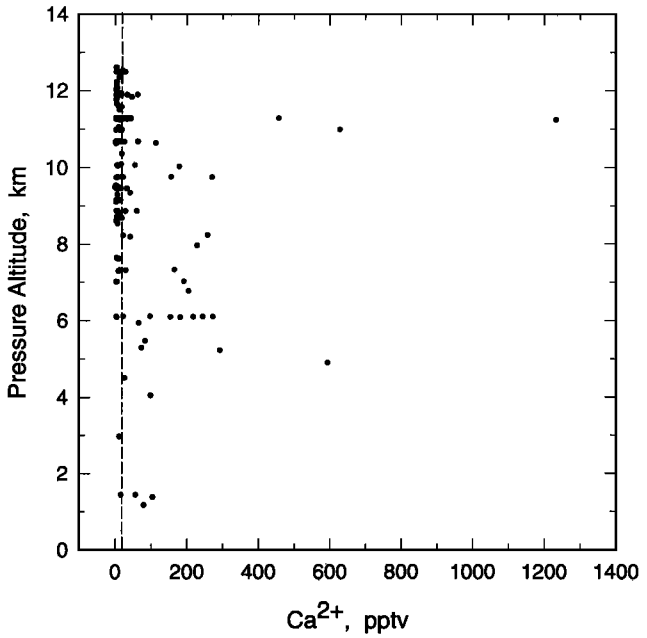

Figure 1. Vertical distribution of aerosol Ca^{2+} over the south-central United States during April-May 1996. The dashed line illustrates the 20 pptv median mixing ratio of Ca^{2+}.

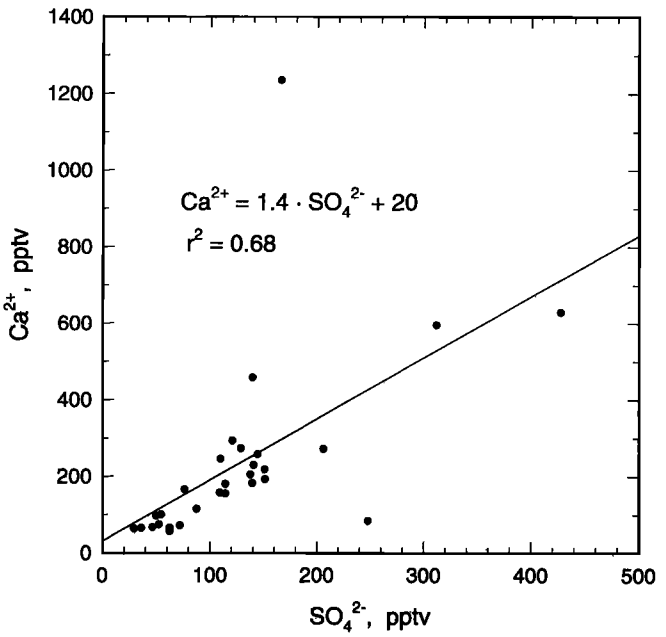

Figure 2. Relationship at 6 - 12 km altitude between mixing ratios of aerosol Ca^{2+} and SO_4^{2-} for Ca^{2+}>50 pptv.

SUCCESS we sampled stratospheric air and never observed evidence for a source of aerosol Ca^{2+}.

Overall, when the mixing ratio of Ca^{2+} was greater than 100 pptv there were also significant amounts of SO_4^{2-}, NO_3^-, and NH_4^+ present in the aerosol. This group of data (n = 25) exhibited median (mean ± one standard deviation) values in pptv of 140 (164 ± 87) for SO_4^{2-}, 134 (163 ± 145) for NO_3^-, and 167 (268 ± 262) for NH_4^+. These values are about a factor of 4 larger than the median mixing ratios observed at 6 - 12 km altitude when Ca^{2+} was <100 pptv: 35 pptv SO_4^{2-}, 40 pptv NO_3^-, and 70 pptv NH_4^+. There was little correlation between NO_3^- and Ca^{2+} ($r^2 < 0.3$) which indicates that NO_3^- was not associated with soil dust as it has been found to be in the boundary layer [*Wolff*, 1983; *Talbot et al.*, 1988].

The best correlation between these three aerosol species and Ca^{2+} was with SO_4^{2-} for mixing ratios of Ca^{2+} >50 pptv (Fig. 2). The most significant outlier point in this correlation occurred at the largest mixing ratio of Ca^{2+} observed during SUCCESS, 1235 pptv. This particular sample was collected at 11.2 km altitude during flight 4 where the DC-8 was profiling through cirrus clouds. The ice particle number density (diameter >4 μm) was as large as 120 cm^{-3} [*Twohy and Gandrud*, this issue]. Since the DC-8 was in ice (cirrus) for approximately 80% of the time covering the aerosol sampling interval, it is likely that we actually sampled ice particles in this instance. These data are consistent with the observation during SUCCESS that ice particles in the middle and upper troposphere commonly contained primarily Ca^{2+} and SO_4^{2-} [*Chen et al.*, this issue; *Rogers et al.*, this issue]. A few other episodes of enhanced aerosol Ca^{2+} on other flights also showed positive correlation with the number density of ice particles. More often, however, the enhanced mixing ratios of Ca^{2+} were observed in clear air.

On a couple of flights we observed positive correlation between mixing ratios of aerosol Ca^{2+} and NO (Fig. 3). In particular, flights 6 and 9 showed this relationship. The correlation between Ca^{2+} and NO is probably related to the transport of boundary layer air enriched in these species to the upper troposphere. It is highly unlikely that such large mixing ratios of NO would persist in long-range transported air parcels. About 50% of the enhanced Ca^{2+} values on these flights were observed in air parcels with ice particle number densities as larger as 400 cm^{-3}, but the rest occurred in apparently clear air. These same air parcels also exhibited positive correlation between the mixing ratio of Ca^{2+} and CO, similar to that shown for Ca^{2+} and NO.

Discussion

The theme of the data observed during SUCCESS suggests a picture of substantial vertical transport of boundary layer air to the

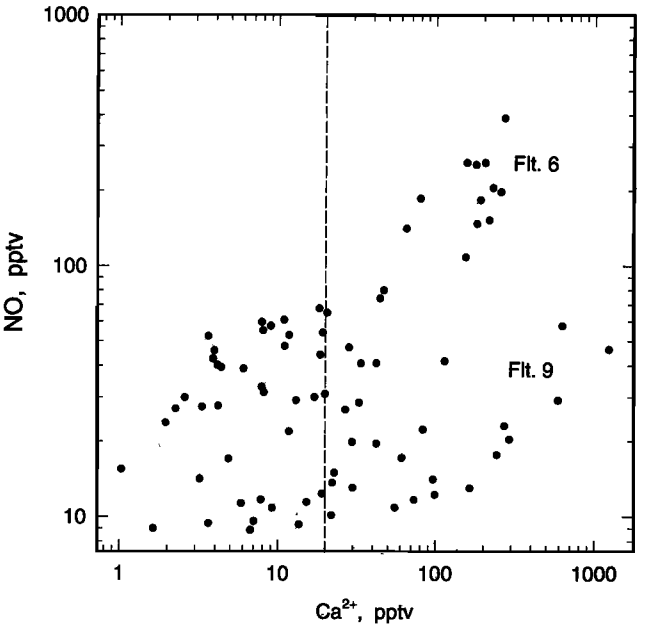

Figure 3. Relationship at 6 - 12 km altitude between mixing ratios of aerosol Ca^{2+} and NO. In particular, flights 6 and 9 over the CART region showed a positive correlation between these two species. The dashed line illustrates the 20 pptv median mixing ratio of Ca^{2+}. Measurements obtained in jet exhaust plumes and contrails (indicated by ppbv levels of NO) are excluded from this plot.

middle and upper troposphere over the central United States during springtime. Indeed, the mixing ratios of aerosol Ca^{2+} in the upper troposphere were among the largest that we have ever measured, but they were consistent with those observed (300 - 600 pptv) in an Asian dust outbreak over the western Pacific during PEM-West B [*Talbot et al.*, 1997]. The unique facet of the SUCCESS data is the extent of the aerosol vertical transport, up to 12 km altitude. In general, long-range transport of crustal aerosols (e.g., Asian and Saharan dusts) has been observed to occur at altitudes of 2-5 km [*Prospero and Carlson*, 1972; *Talbot et al.*, 1986; *Merrill et al.*, 1989; *Perry et al.*, 1997; *Dibb et al.*, 1997].

Summing the individual mass contribution of the measured water-soluble ions provides an estimate of the aerosol (soluble) mass concentration. The median aerosol mass concentration observed above 6 km altitude during SUCCESS was about 0.2 μg m^{-3} STP, but values as large as 6.0 μg m^{-3} STP were found in air parcels seemingly influenced by recent inputs of boundary layer air (i.e., enhanced Ca^{2+}, SO_4^{2-}, NO, and CO). Overall, Ca^{2+} and SO_4^{2-} accounted for 52 ± 28% of the estimated (soluble) aerosol mass concentration.

The measured Ca^{2+} concentrations provide a way to estimate the lower limit of the dust concentrations in the 6 - 12 km altitude range during SUCCESS. Using an average mass concentration of 12,700 μg Ca g^{-1} in world soil [*Bowen*, 1966], the range in dust concentration was 7 - 160 μg m^{-3} STP with a median value of 30 μg m^{-3}. Airborne measurements during dust storms in the early 1970's over the northwestern sections of Texas and Oklahoma showed dust concentrations of 100 - 400 μg m^{-3} STP at 2 - 5 km altitudes [*Gillette et al.*, 1978].

To provide a measure of the influence that vertical transport of boundary layer air to the upper troposphere had on aerosol chemical composition, the sampled air parcels were classified using tracers and geographic location [*Dibb et al.*, this issue]. Four classifications were used: (1) ^7Be concentrations above 1000 fCi m^{-3} STP indicated significant stratospheric influence, (2) samples collected over the eastern Pacific represented upwind source air parcels for the central United States, (3) mixing ratios of Ca^{2+} <100 pptv indicated air parcels without significant continental boundary layer inputs, and (4) mixing ratios of Ca^{2+} ≥100 pptv indicated air parcels with significant continental boundary layer inputs. The resultant aerosol chemistry associated with this scheme was remarkably consistent (Table 1, note that the concentrations are stated in nanoequivalents m^{-3} STP), and it illustrates the substantial impact of vertical transport of boundary layer constituents to the upper troposphere. In addition, the presence of measurable aerosol Ca^{2+} and Mg^{2+} over the eastern Pacific Ocean at this time of year is likely due to long-range transport of Asian dust [*Gao et al.*, 1992].

It is clear from the aerosol compositional summary presented in Table 1 that large mixing ratios of Ca^{2+} were coincident with substantial transport of SO_4^{2-}, NO_3^-, and NH_4^+, and Mg^{2+} into the middle and upper troposphere. This also appears to be true for trace gases such as CO and NO (Fig. 3). There was poor correlation between Ca^{2+} and ^{210}Pb concentrations, presumably due to recent rapid transport of ^{222}Rn gas from the boundary layer to the upper troposphere on time scales too short for significant production of ^{210}Pb in the sampled air parcels.

During the high Ca^{2+} events the water-soluble anion/cation bulk ratio was decreased by a factor of 5 (Table 1). It is interesting to note that the $NH_4^+/(NO_3^- + SO_4^{2-})$ ratio was apparently unaffected. The bulk aerosol compositional ratios of K^+/Ca^{2+}, Mg^{2+}/Ca^{2+}, and Na^+/Ca^{2+} for the convective cases (Table 1) are strikingly similar to those for limestone and/or cement [*Cornille et al.*, 1990]. We have no way of knowing if this apparent limestone-type material represents resuspended soil dust or losses from liming of agricultural fields in preparation for spring crop planting. If the aerosol Ca^{2+} was from limestone, CO_3^{2-} which we did not measure in these samples could account for the approximate 50% deficit in anions for the convective cases (Table 1).

We also compared the S/Ca ratio (i.e., S based on the SO_4^{2-} mixing ratios) in the bulk aerosol convection cases and found that it was on average 10-fold larger than that expected for limestone [*Cornille et al.*, 1990]. This suggests that the majority of the sulfur was derived from anthropogenic sources. Since the aerosol data represent bulk composition, there is no way to determine if the apparent acidic and crustal (limestone) materials were internally or externally mixed [*Andreae et al.*, 1986]. The good correlation between Ca^{2+} and SO_4^{2-} (Fig. 2) could be due to coincident transport of crustal (limestone) material and sulfate aerosol in the same air parcels. Some of the correlation could also be driven by reaction of H_2SO_4 with alkaline crustal material. Support for this reaction is derived from the finding that cirrus nuclei (i.e., crustal material containing Ca) sampled during SUCCESS had a sulfate surface coating [Chen et al., this issue]. Undoubtedly, however, there was probably also vertical transport of boundary layer "anthropogenic submicron aerosol" (i.e., partially ammoniated nitrate and sulfates), as in general sulfate aerosol in the free troposphere over the United States is largely unneutralized [*Ferek et al.*, 1983; *Luria et al.*, 1986].

We can only speculate on the mechanism by which substantial boundary layer aerosols were transported to the free troposphere. It is likely that crustal aerosol was suspended into the atmosphere by wind erosion which commonly occurs during springtime in the southwestern United States [*Gillette and Hanson*, 1989]. In addition, large mesoscale convective complexes (MCC's)

Table 1. Free Tropospheric (≥ 6 km) Aerosol Composition Summary Over the Central USA in Springtime

Air Mass	NO_3^-	SO_4^{2-}	NH_4^+	K^+	Ca^{2+}	Mg^{2+}	Anion/ Cation	$NH_4^+/(NO_3^-+SO_4^{2-})$
Stratospheric (n=43)	1.7 ± 3.0	3.9 ±3.9	3.0 ±3.3	0.36 ±0.70	0.66 ±1.3	0.36 ±0.65	1.4 ±2.1	0.62 ±0.72
Eastern Pacific (n=21)	0.50 ±0.59	2.4 ±2.3	1.7 ±1.8	0.18 ±0.27	1.8 ±2.6	0.33 ±0.39	1.3 ±1.6	0.57 ±0.38
Mid-USA No Convection (n=113)	1.9 ±4.9	2.2 ±2.3	2.3 ±5.7	0.22 ±0.51	0.71 ±0.89	0.41 ±0.60	2.5 ±6.5	0.65 ±0.90
Mid-USA Convective (n=25)	6.2 ±6.2	12.4 ±8.3	10.2 ±11.1	0.42 ±0.62	23.2 ±22.0	3.9 ±2.5	0.51 ±0.15	0.52 ±0.26

Stated values are mean ± one standard deviation in nanoequivalents m^{-3} STP. Ratio values are means of ratio calculated for each sample. n represents the number of data points in each air mass type.

progressed through the CART region periodically during the SUCCESS study period, and some of these were related to severe storm outbreaks. Storm activity due to MCC's tends to be intensified in the south-central and southwestern United States during springtime [*Velasco and Fritsch*, 1987; *Stensrud*, 1996]. Satellite images obtained during SUCCESS indicated that cloud top temperatures in the CART region were typical of the upper troposphere (i.e., 9 - 12 km cloud tops). Some of these were undoubtedly non-precipitating, and boundary layer aerosols may have been transported successfully in convective outflows to the upper troposphere after suspension by strong downdrafts in MCC's or frontal passage activity [*Pye*, 1987]. Whatever the specific transport mechanism was, it occurred routinely as enhanced Ca^{2+}, SO_4^{2-}, NO_3^-, and NH_4^+ was observed at altitudes above 6 km on nearly every flight during the April-May study period. During PEM-West A we sampled convective outflow from continental and marine storm systems and found efficient removal of aerosols compared to insoluble gases such as CO, DMS, PAN, and hydrocarbons [*Dibb et al.*, 1996; *Newell et al.*, 1996; *Talbot et al.*, 1996]. However, these storm systems were predominately associated with wet convective activity. In the spring of 1996 the central United States experienced a modest drought (i.e., precipitation was <50% of the climatological average from January to May), so the environmental conditions favored suspension of crustal materials due to storm activity. We believe that it is unlikely that the crustal aerosol observed in the upper troposphere was related to long-range transport due to the high mixing ratios of dust, NO, CO, and SO_4^{2-} aerosol.

Conclusion

The data presented in this paper indicates that boundary layer aerosols have a significant influence on the chemical composition and mixing ratios of aerosol associated water-soluble species in the middle and upper troposphere over the south-central United States in springtime. Our observations suggest vertical transport of anthropogenic and crustal material in association with storm systems passing through this region. Additional measurements are needed to assess whether this phenomenon was related specifically to the region's drought during 1996 or if it is a common occurrence in spring and summertime. If it is persistent, this vertical transport of aerosol materials could have an impact on cloud and contrail formation over the central United States. It also may have implications for direct (scattering by dust) and indirect (scattering by clouds) radiative effects over continental areas.

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