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The Tucumán solar UV transparency experiment: Preliminary results and prospective

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Two radiometers were simultaneously operated during the past dry season in Tucumán province, NW Argentina. The main objective was to determine particulate matter content of the atmosphere. Ampimpa, an astronomic observatory up in the mountains, was adopted as the reference, non polluted monitoring site. The INTA meteorological facility at Famaillá, in the plains, was chosen as an air pollution test site, its sets of data to be compared to those from Ampimpa. Simultaneous radiometry curves between Ampimpa and Famaillá were almost coincidental in clear days. In cloudy, misty, smoked or rainy days, this feature no longer holds, a fact that is reflected by loss of statistical significance in ANOVA tests performed on corresponding data sets. Using the special geographic characteristics of the region, particulate matter content was calculated by taking air samples and relating them to radiometry data. The experiment also provided the basis for a transparency monitoring network.

Key words: Radiometer, ANOVA, transmittance, particulate matter.

INTRODUCTION

The Tucumán transparency experiment is a ground based simultaneous radiometry experiment designed to obtain atmospheric transparency data for particulate matter (PM) content determination (U.S. Environmental Protection Agency (EPA), 2010a).

Natural and anthropogenic particle emissions in this region reach to its maximum during the dry season, coincidental with late autumn, winter and early spring. The major anthropogenic component is smoke from massive biomass burning, associated with sugar cane crop (Macedo, 1998). Fires cover a good part of the territory, where two producer regions can be distinguished: western, at the foothills and eastern, a dryer zone, but with intense activity (Fandos et al., 2010).

Authors (Amiridis et al., 2009; Chen et al., 2009; Lee et al., 2010; Murayama et al., 2004; Slowick et al., 2007) dealt with similar dust and smoke (PM) conditions with different techniques including light radar (lidar) (Doherty et al., 1999) and satellite radiometry, their main concern being the identification, concentration, lifetime and transport process of dust and smoke (Mulholland and Baum, 1980; Murayama et al., 2004). There are also diseases related to PM in the atmosphere (Goldberg et al., 2001).

The project contemplates the operation of a 5 node radiometer network in order to obtain a good spatial coverage.

The astronomic observatory at Ampimpa (2010) (-26° 48' 03.43", 65°, 50' 36.32" W, elevation: 2458 m) was adopted as a reference site for its good air transparency and location: not beyond 100 km in straight line from the farthest fire. It is the unpolluted reference site, providing control curves and data to be compared to those from other sites (Figure 1).

Atmospheric PM content affects transparency to sun ultraviolet (UV) light (Omar et al., 1999), thus affecting

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Abbreviations: EPA, Environmental Protection Agency; INTA, Instituto Nacional de Tecnología Agropecuaria.



Figure 1. Ampimpa and Famailá monitoring sites.

radiometer irradiation measurements. The quotient between UV irradiation from a clear site and that from a polluted site is proportional to the medium transmittance. The project proposal is to determine PM content from radiometry data and build efficient passive aerosol detection and monitoring networks. This paper shows some preliminary results together with statistical analysis performed on data records, which proved helpful in grouping days with similar transparency and to ascertain Ampimpa as a reference site.

MATERIALS AND METHODS

Three automatic acquisition and recording radiometers were built under the same specifications and components. Photodiode detectors have an imbedded diffuser lens, a feature that makes unnecessary to point the instrument to the sun. They are coupled with amplifiers and A/D converters, and their active spectral sensitivity ranges from 210 to 380 nm. Effective ground based

detection starts at about 280 nm.

Three sets of data were obtained from three different sites during the 2010 campaign: Argentina's National Institute for Agricultural Technology (INTA, 2010) meteorological facility at Famailá ($-27^{\circ} 01' 08.85''$, $65^{\circ} 22' 49.99''$ W, elevation 373 m) during September (Figures 2a and 2b) and October (Figures 2c and 2d), city of Tucumán ($-26^{\circ} 49' 17.68''$, $65^{\circ} 11' 37.98''$ W, elevation 441 m) during the test period in August (Figure 3a), and the astronomic observatory at Ampimpa, during October, (Figure 3b). Data and figures in this paper were referred to DST (Daylight Saving Time) = UT - 3, one hour ahead of LT.

The radiometer at Ampimpa was the master instrument; all others were calibrated to fit its signal readings within a 20 mV tolerance.

By August, sugar cane harvest activity was declining to an early end and fires became sparser and sparser, until they finally ceased, at the end of October. Diurnal data records spanned between 09:00 DST and 19:00 DST. Records included cloudy, polluted and clear, non polluted days, which were used for comparisons to polluted days.

A statistical analysis was performed to detect similarities among records, and to ascertain Ampimpa as an adequate reference site.

Similarities among records were initially explored by cluster

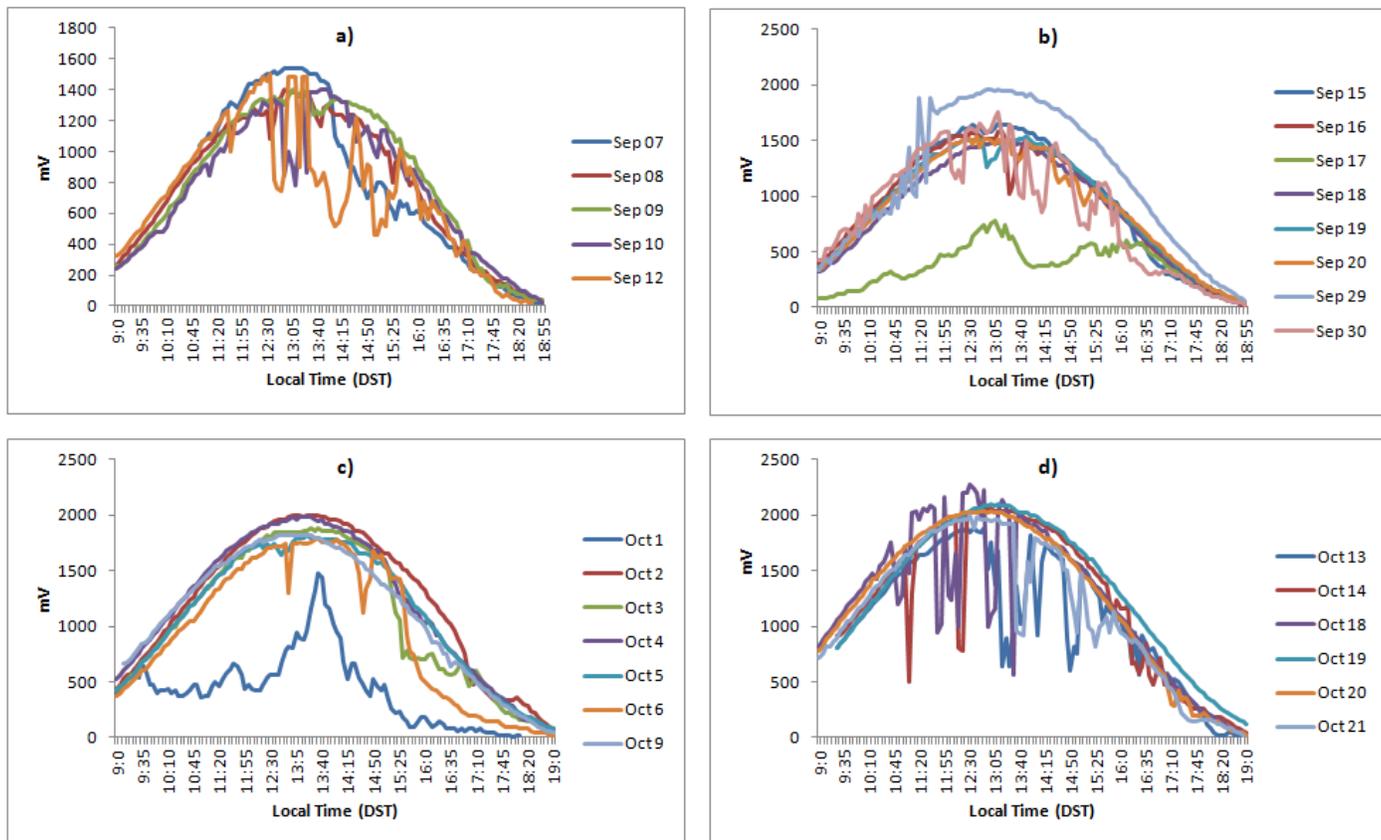


Figure 2. Raw curves for Famailá.

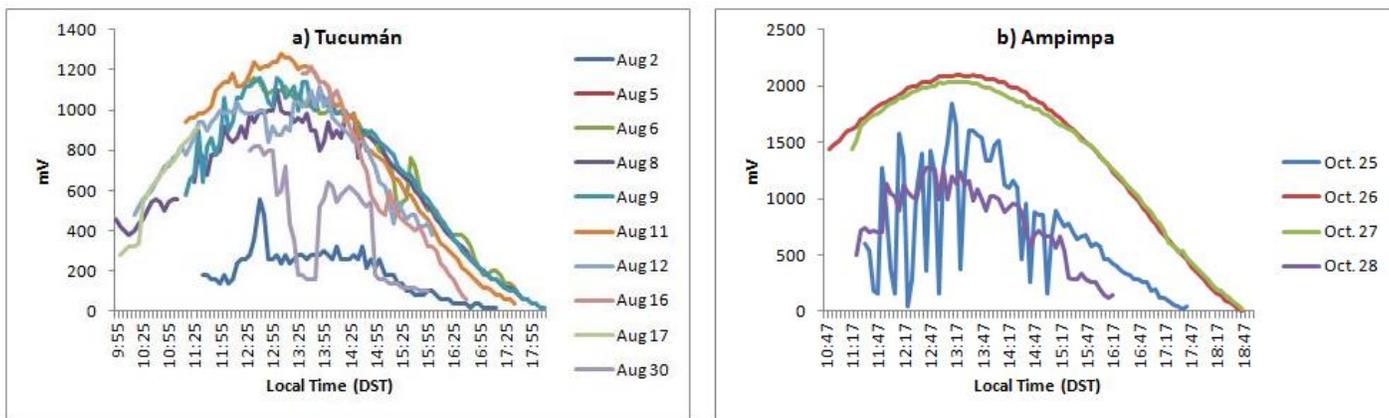


Figure 3. Raw curves for (a) Tucumán and (b) Ampimpa.

analysis (Hartigan, 1975; Kaufman and Rousseeuw, 1990). In this technique, sets of records are assigned into groups (called clusters), so that the records in the same cluster are more similar to each other than to those in other clusters. Similarities were taken in the sense of models based on distance connectivity (hierarchical clustering). Euclidian distance was the criterion adopted since the interest was centered on seeing any difference in transparency among days based on how close is a point to its

neighbor. This criterion is based on the core idea of objects being more related to nearby objects than to objects farther away.

Statistical inference analysis was also performed on data records. Each day is considered a treatment and an ANOVA test explored differences among treatments (Maxwell and Delaney, 2004; Keppel and Wickens, 2004). It was used to establish statistical significant differences among data records. Differences were encountered, but this technique does not establish differences

among precise days. A Tukey test was essayed (NIST/SEMATECH, 2010) to find which records were significantly different from one another, by comparing all possible pairs of record means. This method considers all possible pairwise differences of means at the same time.

PM content determination can be achieved starting from Beer's law which, for an atmospheric layer, can be written as:

$$I = I_0 e^{-\tau} \quad (1)$$

I_0 is the incident radiation, I the radiation exiting the layer and τ the optical depth of the layer. The layer transmittance is:

$$T = \frac{I}{I_0} = e^{-\tau} = e^{-\sigma d N} \quad (2)$$

d is the airmass (path traversed by sun radiation in the layer), σ the absorption cross-section (m^2) and N the number density of absorbers (molecules $\times \text{m}^{-3}$), with l the unit of longitude. Further on

$$\ln(T) = \ln\left(\frac{I}{I_0}\right) = -\sigma N d \quad (3)$$

In Equation (3), the dimensionless exponent is composed of particle cross-section σ [m^2], airmass d [m] and numerical density [number of particles per m^3]. The already available quantities are transparency and ground level PM mass density estimate. Also, airmass concept differs, since it is not related to the light path through the atmosphere but to a small portion, its length estimated as:

$$l = (h_{\text{Ampimpa}} - h_{\text{Famaillá}}) / \cos(z)$$

Here h_{Ampimpa} and $h_{\text{Famaillá}}$ indicate the altitudes of Ampimpa and Famaillá, respectively. If the starting point is equation (3), replacing $\sigma N d$ with the column weight W_T :

$$\ln(T) = -\sigma N d = -K W_T \quad (4);$$

N is linearly related to W_T . Then:

$$K = -\ln(T) / W_T \quad (5);$$

K depends on geometry (d), and the type of pollutant (σ) which we assume the same throughout the experiment. For a given K , Equation (4) yields an estimate of the total pollutant weight W_T along the light path between the altitudes considered.

RESULTS

Statistical analysis

Cluster analysis (average linkage object grouping) was employed as the exploratory data analysis technique. Clusters can be represented by tree diagrams such as those displayed in Figure 4.

In Figure 4a, day 17 did not cluster to the rest, while day 18 and day 30 were almost identical (Figure 2b). Day17 exhibited the least radiance. Day 12 was cloudy,

Its curve strikingly different from those of days 16 and 19, with significant reduction in radiance levels, whilst September 16th, was a bright day. Day 29 was another dissimilar day, some mist in the morning hours and very bright and sunny starting at 11:30 am.

An ANOVA of September data set for Famaillá resulted in $F = 22.57$ with $p < 0.0001$, which meant statistically significant differences among data records. A Tukey test was performed to detect those differences among precise days (Table 1). In all Tukey tests, significance level α was set to 0.05.

In Table 1, the calculated minimum significant difference (MSD) value was 192.84637. Difference between means larger than MSD indicates statistical significance. Day 17 (cloudy) was statistically different from the rest of the days included, as indicated by letter A in the fourth column.

Day 29 was another singular day, bright and sunny with no fires; it began somewhat misty but cleared at noon, with higher irradiation values (Figure 2b). Day 12 was cloudy, and shared some statistical similarity to days 8, 9, 10, 18 and 30 (days with fires), but not to days 16, 17, 19, 20 and 29.

In Figure 4b, October 1st, was a cloudy day which appeared very different from the rest. Data clustering joined day 2 (a clear day with some smoke at about 16:00 DST) with day 18, a bright day with smoke detection between 10:30 DST to 15:00 DST, and rendered day 9 as identical to day 5. Also, similarities between days 3 and 13 and between days 4 and 14 were readily detected. October 6th is another dissimilar day. It was a clear day, with no fires, although a few clouds were detected at 13:00 and 14:30 DST.

ANOVA on Famaillá, October data set, resulted in $F=14.93$ with $p < 0.0001$, an indication of statistically significant differences among records. Tukey test (MSD = 267.22118) in Table 2 indicated statistical significant differences between day 1 and each one of the remaining records, and also between day 6 and days 2, 18, 19 and 20.

October 1st differences were so striking that they could have influenced on the analysis, so a new ANOVA was performed after taking this record out. Although F value decreased to 2.81 with $p < 0.0012$, the general behavior persisted. A new Tukey test (Table 3) again found significant differences between day 6 and each one of days 2, 18, 19 and 20, which, in turn, were similar among themselves (MSD = 269.83273).

In order to determine the adequacy of Ampimpa as a reference site, an ANOVA was conducted on Ampimpa, day 26 and Famaillá clear day records (days 19 and 20 in Figure 5a). Here $F = 2.17$ with $p < 0.1163$ meaning there were no statistically significant differences among records in clear days. In Figure 5b), the hourly variations of raw data for Tucumán (08/11/2010); Famaillá (09/18/2010 and 10/19/2010) and Ampimpa (10/26/2010) are plotted. Note the similarity between Famaillá and Ampimpa curves.

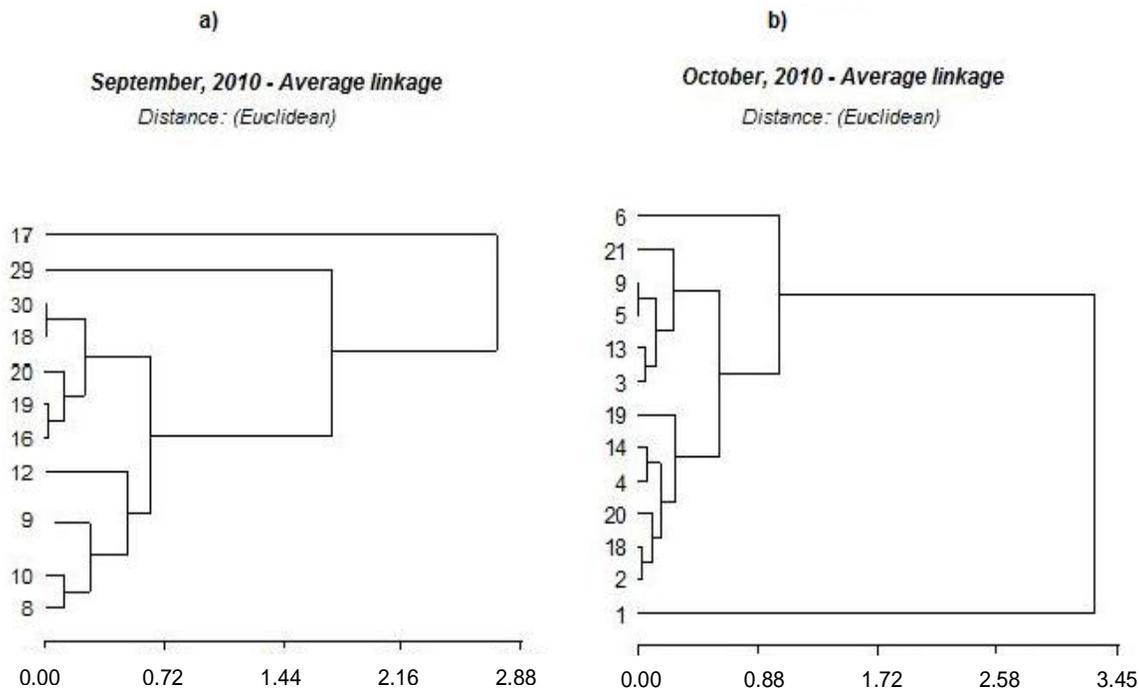


Figure 4. Cluster tree diagrams of Famaillá data records, a) September and b) October, 2010.

Table 1. Tukey test for Famaillá, September 2010.

September days	Mean	n			
17	384.27	117	A		
12	733.68	117		B	
10	792.14	117		B	C
8	812.82	117		B	C
9	846.32	117		B	C
18	898.46	117		B	C
30	900.17	117		B	C
20	929.23	117			C
19	949.74	117			C
16	954.87	117			C
29	1192.99	117			D

Different letters indicate significant difference ($p \leq 0.05$).

Particulate matter content estimation

Equation 4 states that it is possible to determine W_T and hence, the numerical density of PM from Ampimpa and Famaillá radiometry data records. Particle mass density at ground level was initially determined by air sampling on October 21st, 15:00 DST (Figure 2d). On this particular day, the atmosphere was clear until noon (DST) when a fire started in a nearby field, very close to and SE of the radiometer site. Soon after, smoke reached to the detector; the plume height was estimated in 200 m above

ground level, in a scenario similar to that depicted in Figure 6.

Air samples were taken sucking in through a 0.45 μ paper filter with a vacuum pump. Dry filters were weighted twice: before and after sampling; the weight difference, 0.0048 g, was attributed to the accumulated PM. The aspired air volume was estimated in 18 L, which results in a PM mass density estimate of 266.7 $mg \times m^{-3}$ at ground level.

Rough PM content estimates for the vertical path, based only on the fraction of atmosphere below an

Table 2. Tukey test for Famaillá, October, 2010.

October days	Mean	n			
1	443.60	111	A		
6	960.67	120		B	
3	1080.33	120		B	C
13	1091.75	114		B	C
9	1116.17	120		B	C
5	1117.50	120		B	C
21	1158.07	119		B	C
4	1202.81	121		B	C
14	1218.31	118		B	C
2	1235.50	120			C
18	1242.69	119			C
20	1262.67	120			C
19	1290.91	121			C

Different letters indicate significant difference ($p \leq 0.05$).

Table 3. Tukey test for Famaillá, October 2010, day 1 excluded.

October days	Mean	n			
19	1290.91	121	A		
20	1262.67	120	A		
18	1242.69	119	A		
2	1235.50	120	A		
14	1218.31	118	A	B	
4	1202.81	121	A	B	
21	1158.07	119	A	B	
5	1117.50	120	A	B	
9	1116.17	120	A	B	
13	1091.75	114	A	B	
3	1080.33	120	A	B	
6	960.67	120		B	

Different letters indicate significant difference ($p \leq 0.05$).

altitude of 650 m above sea level, were obtained by relating values stored in Table 4.

Transmittances were calculated for the slant path traveled by sun rays between the altitudes of Famaillá and 200 m above ground level. The table was built under the assumption of dry air, so that PM suspension capability varies according to the fraction of atmosphere below. In column 4, summation of partial mass content yields a total PM weight estimate of 65.23 g for this 200 m vertical column. The calculated vertical content value must be divided by $\cos(z)$ for the slant path, yielding a total slant content of 76.32 g. At that time (15:00 DST) atmospheric pressure in Famaillá was 1005.6 hPa, solar zenith angle $z = 31.28^\circ$, $I_0 = 1700$ and $I = 1320$.

From Equations (3) and (5): $-\ln(T) = -\ln(1320/1700) = 0.25299651$, $W_T = 76.32$ g; resulting in $K = 0.0033$ g⁻¹.

As an example, consider the low radiometry value (920 mV) measured the same day, at 14:05 DST. The corresponding Famaillá October 19 (control curve) radiometry datum was 1980 mV; then PM total content along the slant path between the altitudes of Famaillá and 200 m above ground level is:

$$W_T = -\ln(T)/K = -\ln(I/I_0)/K = -\frac{\ln(920/1980)}{0.0033} = 232.27 \text{ g}$$

At 14:05 DST, $z = 21.38^\circ$; then the calculated vertical content is 216.3 g. Taking this target value and adjusting the calculation process using the second column in Table 4, yields a ground level PM density value of 672.5 mgxm⁻³. Calculations based on this approach will hold as long

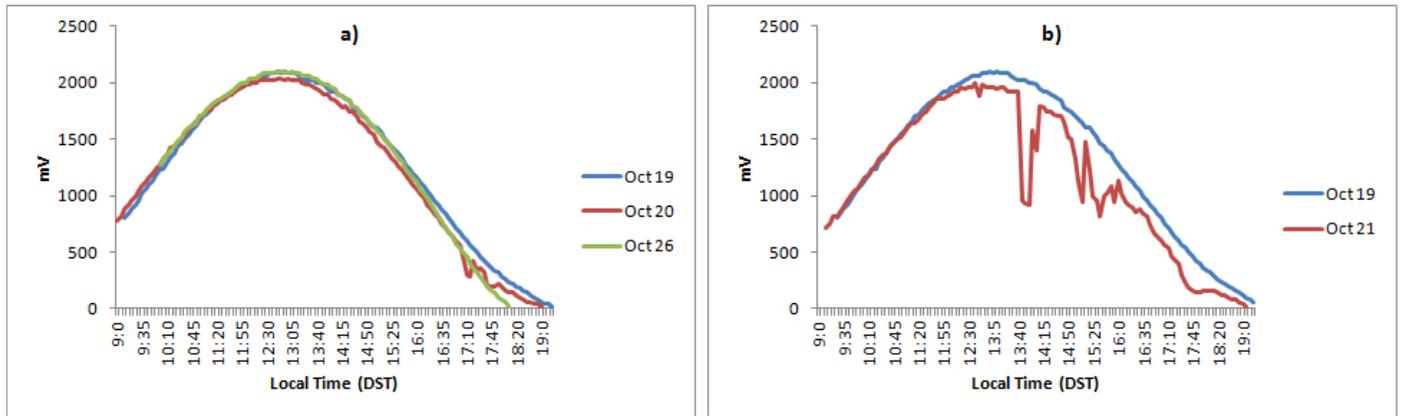


Figure 5. Clear days in October, 2010. (a) Famaillá days 19 and 20 as compared with Ampimpa, day 26 and (b) Famaillá day 19 (a clear, reference day) with day 21, clear, but with nearby fire.

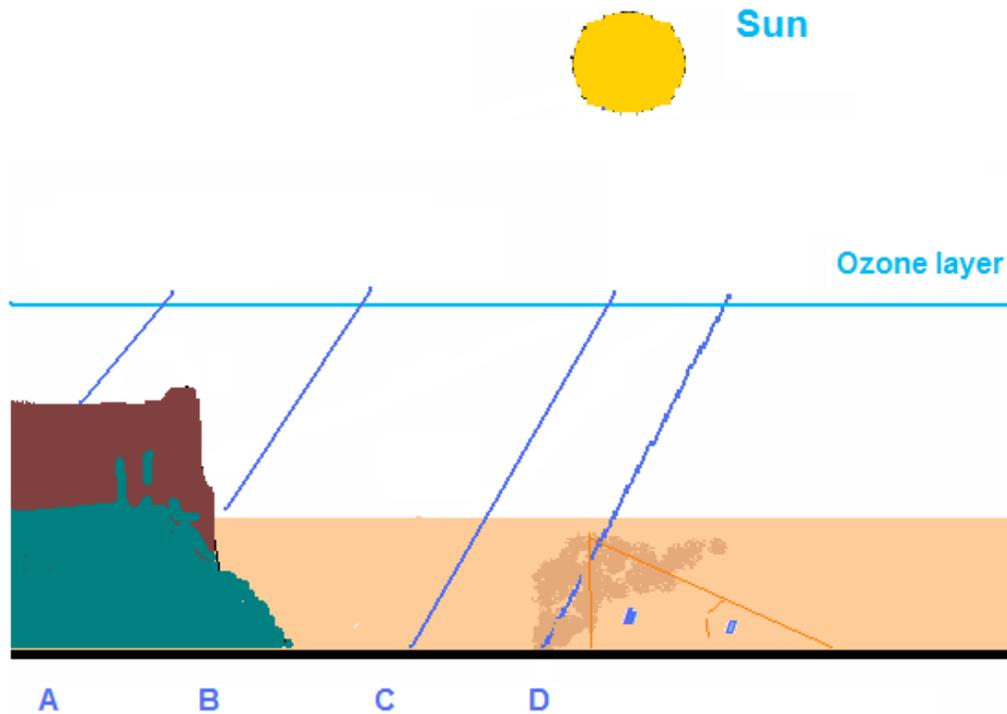


Figure 6. A sketch of proposed sun radiometer deployment sites and smoke plume detection: A, Ampimpa; B, San Javier; C, Famaillá; D, smoke plume site.

as zenith angles remain less than 60°.

Measured and calculated density values exceed the standards for the referred type of PM, according to the EPA National Ambient Air Quality Standards for Particle Pollution table (2010b).

DISCUSSION

Solar irradiation varies with the inverse square of Sun-

Earth distance, the difference that arises because of Earth’s orbital position is about 1.5% and is not significant for the present purposes. Another source of fluctuation in results is the sun zenith angle, with pronounced variation between epochs of the year. This variation is reflected by peak values in the irradiation curves: the closer to summer solstice, the higher they grow (Figure 7). Here, curves for Tucumán (August 11th) and Famaillá (September 9th) exhibit the effects of pollution. Figure 7 also displays the Famaillá, October

Table 4. PM density and weight vs. altitude above Famaillá. Relative pressure values were interpolated from the 1976 U.S. Standard Atmosphere (PDAS, 2010); the fourth column results from multiplying the third column density values by the adopted height interval (25 m) times the square meter section.

h (km)	P/P_0	PM density ($\text{g}\times\text{m}^{-3}$)	PM vert. content (g)
0.4	0.96068	0.26667	5.123690712
0.42	0.96022	0.26654	5.118740776
0.44	0.95833	0.26601	5.098507266
0.46	0.95644	0.26549	5.078505112
0.48	0.95456	0.26496	5.058404352
0.5	0.95267	0.26444	5.038481096
0.52	0.95079	0.26392	5.018649936
0.54	0.9489	0.26339	4.99861542
0.56	0.94702	0.26287	4.97862948
0.58	0.94513	0.26235	4.95909711
0.6	0.94325	0.26183	4.93942295
0.62	0.94137	0.2613	4.91959962
0.64	0.93995	0.26078	4.90240322

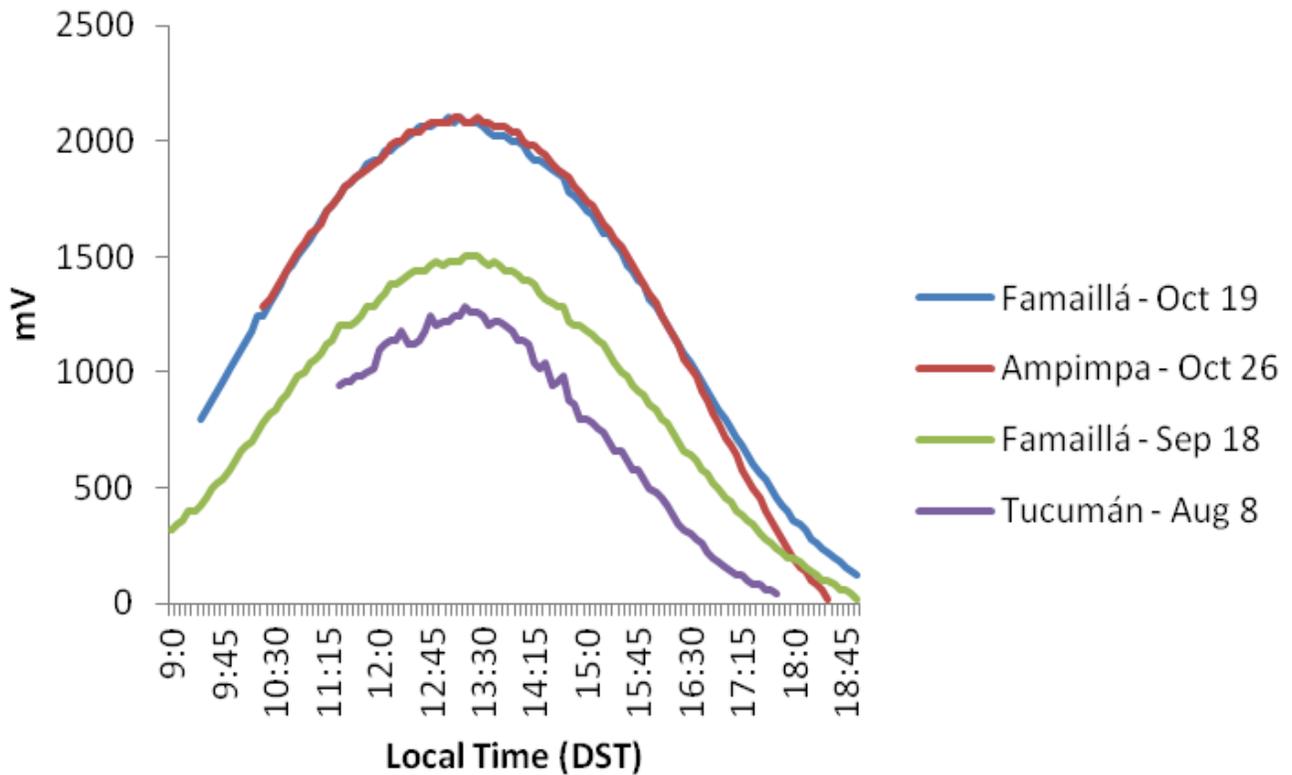


Figure 7. Seasonal effect on radiometer curves.

19th and Ampimpa, October 26th clear day records with almost no difference between them, a further evidence of Ampimpa as a provider of good control curves.

Statistical analysis reflects grouping in radiance data: ANOVA results confirm the general picture attained

through cluster analysis. It is also an indication of the dispersion of irradiance values. Further correspondence among records from sun irradiance standpoint was determined after a Tukey test.

A more definite conclusion on how data group, and how

many groups are distinguishable, can be reached to only after the inclusion of many days, so a new campaign is being planned for the dry season in 2011. Anyway, statistics is used on radiance data and, so far, cannot make distinction among misty, cloudy, smoked or dusty days. Sunny, bright days with no fires are easily separated from the rest, and so do thickly cloudy or rainy days. There were no densely smoked days, mostly because Ampimpa and Famaillá radiometers were operated after the sugar cane crop climax, and instruments were picking disperse fire smoke. Should data were collected during the months of July and August, still under intense industrial activity and burning, records would have reflected a thicker, homogeneous, highly polluted atmosphere. A more precise K value would have been attained if PM initial density determinations were carried out in such an atmosphere. In this case, the limiting altitudes of airmass are those of Famaillá and Ampimpa. These conditions did not occur during the 2010 recording period.

In their paper authors Murayama et al. (2004) referred that Åmströng exponent and extinction coefficient bulge at two different altitudes below 5 km altitude. Chen et al. (2009) reported extinction coefficient bulges in their profiles at altitudes of about 500 and 1250 m, a reference that can be helpful in understanding what the PM density profile might look like above Tucumán plains. There is, nevertheless a difference: most Asian results refer to not so fresh aerosol molecules, while Tucumán is a small province, a soot producing source, and is next to (and overlaps) dust producing regions (Figure 1).

Slowik et al. (2007) have demonstrated that recently emitted soot particles are typically fractal aggregates. Black carbon (BC) spherules absorb light individually and the total light absorption is proportional to the mass of the fractal particle. Also, fractal condition changes with time, evolving to compact configurations, a concept that cannot be extended to the present case because of the shorter time periods involved. Now, sugar cane leaves burn down to different sizes, hard ashes called "malhoja" (bad leaf), ranging from a few millimeters to several centimeters long, with no estimated media. If they are fractal or not, this is something that has not been established yet. Ashes are raised by combustion hot air and carried by the wind. Though these ashes do not make up of a major abundance component, as compared with other combustion products, they do absorb light and can also sting eyes, with consequences most of the times.

Regarding this, an interesting location is on top of San Javier hill (-26° 47' 56.88", 65° 21' 40.45" W, elevation: 1272 m), overlooking the city of San Miguel de Tucumán: it has a clearer atmosphere, with a good view of most of the pollution above the plains. The idea is to compare the transparency below this altitude and between this altitude and Ampimpa. Yet there may be some soot generated in the plains above this altitude, and of course, is going to

be dust in suspension; but the point here is to evaluate the relative abundance of PM between these altitudes and its contribution to transport processes.

The proposed sampling method for PM content estimation stems on the validity of Equation (3), which carries implicitly the admission of a formal dependence of transparency on pollutant mass. If a mass content can be determined and its variation with altitude calculated, it is straightforward to relate PM content to transparency. Fires are responsible for smoke and also for greenhouse gases, which, in turn, increase atmospheric temperature and therefore, its dust and smoke suspension capability. Thus, fires can also be blamed for more dust. Dust acts mainly by scattering UV light and smoke by absorption (Shiple et al., 1983), so light extinction can be attributed to both of them.

Conclusions

The method adopted for particulate matter content determination makes – so far – no distinction between dust and smoke. It is effective, nevertheless, in estimating PM content. Since PM total content is linearly related to the calculated layer transmittance; K parameter calculation should be carried out once a thick, stable, polluted atmosphere is reached. Also, other paper filter sizes are needed; specifically, 2.5 μ and 10 μ filters should be used in order to determine $PM_{2.5}$ and PM_{10} contents.

Extinction of UV solar radiation is the only reason for lack of statistical significance between Ampimpa and Famaillá simultaneous records, and also among Famaillá records. On polluted days, since fires start mostly at noon (LT), similar raw data curves can be expected.

The statistical analysis supported the following conclusions: (1) there are no significant differences on clear days between Famaillá and Ampimpa; (2) there are significant differences between them when fires are active; (3) the analysis also rendered Ampimpa as an adequate reference site; (4) for the same site, no significant differences appear among daily records when fires are burning: this situation reverses in case of thickly cloudy or rainy days and also when a clear, unpolluted day happens.

Since particulate matter transport is another research area of interest, it becomes necessary to operate at least one more radiometer in the plains. There are plans to widen the radiometry network, specifically to include San Javier hilltop facilities as recording site. Equipments must be inter-calibrated when there is no pollution and humidity is low, preferably in early autumn. Radiometry data should be also used in connection with satellite data for the area, MODIS (2011) or the like.

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