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Referência Completa

Tipo da Referência	Conference Proceedings
Repositório	sid.inpe.br/iris@1905/2005/07.26.21.36
Metadados	sid.inpe.br/iris@1905/2005/07.26.21.36.05
Site	mtc-m05.sid.inpe.br
Rótulo	2670
Chave Secundária	INPE-3916-PRE/
Chave de Citação	KirchhoffNobr:1986:OzMeNa
Autor	1 Kirchhoff, Volker Walter Johann Heinrich 2 Nobre, Carlos Afonso
Grupo	1 DGE-INPE-BR 2 CPT-INPE-BR
Título	Atmospheric chemistry research in Brazil: ozone measurements at Natal,Manaus, and Cuiaba
Nome do Evento	Meeting of the Geophysics Comission IPGH.
Ano	1986
Data	7-11 abr. 1986
Palavras-Chave	GEOFISICA ESPACIAL, BRASILNATAL(RN)MANAUS(MA)cuiaba, OZONIO, medição.
Idioma	En
Tipo Secundário	PRE CI
Area	CEA
Projeto	OZONIO
Ultima Atualização dos Metadados	2015:04.16.13.49.47 sid.inpe.br/bibdigital@80/2006/04.07.15.50 administrator
Estágio do Documento	concluido
e-Mail (login)	marciana
Grupo de Usuários	administrator
Visibilidade	shown
Transferível	1
Tipo do Conteudo	External Contribution
Unidade Imediatamente Superior	8JMKD3MGPCW/3EU29DP 8JMKD3MGPCW/3EUPEJL
Conteúdo da Pasta source	não têm arquivos
Conteúdo da Pasta agreement	não têm arquivos
Histórico	2015-04-16 13:49:47 :: administrator -> marciana :: 1986
Campos Vazios	abstract accessionnumber affiliation archivingpolicy archivist booktitle callnumber conferencelocation copyholder copyright creatorhistory descriptionlevel dissemination documentstage doi e-mailaddress edition editor electronicmailaddress format isbn issn lineage mark mirrorrepository nextedition notes numberoffiles numberofvolumes organization pages parameterlist parentrepositories previousedition progress publisher publisheraddress readergroup readergroup readpermission resumeid rightsholder secondarydate secondarymark serieseditor session shorttitle size sponsor subject targetfile tertiarymark tertiarytype type url versiontype volume
Data de Acesso	30 jul. 2015

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Fechar

1. Publication Nº: INPE-3916-PRE/951

2. Version:

3. Date: Jun, 1986

5. Distribution: ☐ Internal ☒ External ☐ Restricted

4. Origin: DGA/DGN — Program: OZÔNIO

6. Key words - selected by the author(s)
OZONE
ATMOSPHERIC CHEMISTRY
SURFACE OZONE

7. U.D.C.: 523.4-836-36

8. Title — INPE-3916-PRE/951

ATMOSPHERIC CHEMISTRY RESEARCH IN BRAZIL: OZONE MEASUREMENTS AT NATAL, MANAUS, AND CUIABÁ

10. Nº of pages: 23

11. Last page: 20

12. Revised by

Enio B. Pereira

9. Authorship V.W.J.H. Kirchhoff
C.A. Nobre

Responsible author

13. Authorized by

Marco Antonio Raupp
Director General

14. Abstract/Notes

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15. Remarks *This work was presented at Meeting of the Geophysics Comission, IPGH, Rio de Janeiro, April 7-11,1986. This work was partially sypported by the "Fundo Nacional de Desenvolvimento Científico e Tecnológico" under contract FINEP 537/CT.*

ATMOSPHERIC CHEMISTRY RESEARCH IN BRAZIL: OZONE MEASUREMENTS AT NATAL, MANAUS, AND CUIABÁ

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Meeting of the Geophysics Comission, Instituto Panamericano de Geografia e História, IPGH, Rio de Janeiro, April 7-11, 1986.

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ABSTRACT

Ozone measurements have been performed in three very distinct ecological environments in order to evaluate regional influences at the surface on the lower atmosphere. Natal (6S, 35W) is a marine station where ozone measurements have been made since 1978 using balloon-sondes as well as rocket-sondes. More recently, measurements have also been made in an equatorial rain-forest environment, in Manaus (3S, 60W) and in the area of the Brazilian savannah, Cuiabá (16S, 57W). It is found that the diurnal variation of the ozone concentration has important distinctions in these different regions. In the forest, O_3 concentrations are very low in comparison to results of the savannah, and specially in comparing with city areas where anthropogenic activities increase the level of atmospheric ozone. Despite being a marine station, which generally shows low ozone characteristics, Natal in springtime has ozone concentrations comparable to Europe and the U.S. The cause for this behavior appears to be related to large scale circulation of the atmosphere. Natal is close to a large subsidence area over the South Atlantic. It is argued that the downward motion associated to the Hadley-Walker circulation cells can direct air over Natal, air that was polluted by agricultural biomass burning in the savannah regions of central Brazil and equatorial Africa.

INTRODUCTION

Among the various gaseous compounds of the atmosphere, specially those that are much less concentrated than the major oxygen and nitrogen molecules, ozone stands out owing to its role in absorbing damaging ultraviolet light. This absorption of UV, or shielding effect, takes place in the stratosphere. A minimum of ozone is therefore required, in the stratosphere, to perform the shielding effect. While in the stratosphere ozone is destroyed by combustion products, or pollution gases, in the lower troposphere the opposite may occur, that is, an increase of ozone results in areas of industrial activity. This increase in ambient ozone concentrations may cause serious inconveniences, if one recalls that ozone is a toxic gas when its concentration increases above certain limits(about 100 ppbv). For example, ozone seems to be responsible for crop damage during severe pollution events. Field studies by Heck et al. (1982) show that crop production decreases at a rate of about 7% per 10 ppbv ozone increase.

Besides the practical aspects of ozone research, the O_3 molecule, being very reactive, takes place in a large number of chemical reactions, and is, therefore, very interesting as well in an Atmospheric Chemistry sense. The atmospheric reactive constituents, that is, those that take part in chemical transformations under the driving force of solar energy, are to some extent, as yet not well-known, influenced by the ecological environment of the surface. Different kinds of vegetation emanate different kinds of gases, in different amounts, to the atmosphere. Different environments represent different sources or sinks for given atmospheric gases.

The map of Brazil shown in Figure 1 shows the different kinds of vegetation that cover the country. We are specially interested in the equatorial rainforest, which is known to contribute a large number of hydrocarbons, mainly isoprene, to the lower atmosphere (Zimmermann et al., 1978), which might represent precursors for ozone production. The large areas of wet lands, along the margins of the

Amazon river and its tributaries, contribute to important amounts of methane to the atmosphere (Harriss, private communication). Another important environment for interaction with the lower atmosphere is the savannah (campo cerrado) region of central Brazil. This large area of short bushes and grassland is put on fire regularly in the dry season and this contributes significantly to several pollution gases to the lower atmosphere (Crutzen, 1979; Crutzen et al., 1985). A semi-arid area in the northeast of Brazil (caatinga) has its own characteristics, not yet explored. Finally, the wetlands of the pantanal, along the Paraguai river,may have a strong impact on the chemistry of the lower atmosphere, but remains unexplored as well. Monitoring stations are shown by heavy dots on the map. Extensive measurements have been made in Natal; monitoring at Manaus and Cuiabá has started only recently.

It should also be noted, that Atmospheric Chemistry in Brazil has a great potential not only because of the richness of its ecological environments, but also because it is close to the equatorial belt where the solar energy input is largest, specially in the more energetic UV spectral range. A lot of energy is, therefore, available to drive chemical reactions in the lower atmosphere. Finally, because of the large extension of the equatorial forest, local sources or sinks for atmospheric gases may have global importance as well.

The aim of this paper is to quickly review results of O_3 measurements in Brazil and to discuss transport effects, specially those related to the Walker cell atmospheric circulation, dry convection produced by large vegetation fires, and direct transport of pollution gases from the African savannah.

RESULTS

In what follows we discuss diurnal and seasonal variations.

Diurnal Variations

The typical shape of the diurnal variation observed for surface O_3 concentrations at Brazilian stations is shown in Figure 2. Ozone concentrations are larger in the daytime. The time of maximum concentration, (T. Max.), varies between about noon and 14 hours local time (L.T.), whereas at night concentrations are smaller, reaching a minimum at T. Min. of about 3 L.T.. The amplitude is generally about 15 ppbv.There is some day to day variation in these parameters, and sporadic drastic changes in their values may occur. The largest difference between stations is observed in the diurnal average, (D.Av.). This parameter is close to zero ppbv in the forest area of Manaus, D.Av. is about 12 ppbv in the cerrado, and between 15 and 20 ppbv in the downtown area of São José dos Campos.

The observed shape of the diurnal variation is similar to observations of the Northern Hemisphere (Fehsenfeld et al., 1983) but different from results obtained in the Pacific (Oltmans, 1981) where the lower atmosphere has less influence from natural (vegetation) and anthropogenic (combustion) gaseous contributions, as well as very different meteorological aspects.

Short term ozone concentration variations may occur from three distinct contributions; injections from upper air richer in ozone, horizontal transport, or local photochemical production. The long term relatively steady contribution of ozone that counterbalances the continuous loss at the surface is a downward flux from the lower stratosphere. In the absence of horizontal irregularities it is easy, therefore, to understand the nocturnal minimum. In the afternoon a stable air surface layer develops which inhibits mixing. The ozone in this layer is consumed at the ground and is not replaced by mixing. The concentration, therefore, decreases during the night.

With sunrise and heating of the surface, the stable layer is disrupted, the mixing layer starts to grow and ozone from ever higher levels is mixed into the mixing layer such that there should be a tendency for the O_3 concentration to increase. More difficult to explain in the diurnal variation cycle is the early afternoon decrease of ozone concentration. We have discussed this part of the variation arguing in favor of local photochemistry. For details see Kirchhoff (1986).

Seasonal Variations

Until recently it was thought that the tropics had much less ozone than the midlatitudes (Galbally and Roy, 1980) or that the Southern tropics had much less ozone than the Northern tropics (Fishman et al., 1979). This view has been completely changed with the results obtained at Natal (Kirchhoff, 1984; Logan, 1986; Kirchhoff and Logan, 1985; Logan and Kirchhoff, 1986). Natal is presently the only tropical station where regular ozone measurements are performed in the troposphere and stratosphere, and it has the largest collection of ozone data obtained at low latitudes. Early results have been described in Kirchhoff et al. (1981, 1983). The seasonal variations in the troposphere have been discussed in Logan and Kirchhoff (1986).

Ozone concentrations in the whole troposphere of Natal show a very strong seasonal variation. In Figure 3 we show ozone concentrations in terms of a percentage variation about the mean at given heights (pressure levels). It is quite clear that a strong seasonal oscillation is present. There is a factor of two between the April-May minimum and the September-October maximum. With increasing altitudes this factor decreases, being practically absent in the stratosphere. It is interesting to note that the absolute values of the Natal spring period are comparable to European and U.S. concentrations (Logan, 1986).

DISCUSSION

It seems reasonable to blame the spring maxima of ozone concentrations at Natal to biomass burnings of the cerrado. Large ozone concentrations in this savannah region of central Brazil have been attributed to biomass burning events (Crutzen et al., 1985). Burnings can contribute to several gases to the atmosphere and these can indirectly produce ozone in a large chemical reaction chain (Crutzen, 1979). However, the prevailing winds at Natal are easterlies in the whole mixing layer, and thus any possible transport of pollutants from the cerrado burnings is not direct but must occur in a more complex scheme.

Natal is close to a large subsidence area. For air from the cerrado to reach Natal it must first ascend to higher levels above about 400 mb, where the prevailing wind regime is eastward (Logan and Kirchhoff, 1986). It appears that two mechanisms could make the air of the cerrado reach the upper levels of the troposphere: direct dry "convection" cells (thermals) produced through the heat generated by the fires, or through the ascending branch of the Walker cell. In addition to the influences of the cerrado burnings we argue that identical burning episodes occur in Southern Africa, which could also contribute significantly to "polluted" air observed at Natal. In what follows we examine these three possibilities.

The campo cerrado region of central Brazil is relatively large and it is not rare that significant areas, up to 50 to 100 km in diameter, are set on fire. The heat produced in these fires is likely to start a localized convection cell with the warmer air of the surface ascending through the lower troposphere. Such a process could transport a considerable amount of gases, ozone precursors included, to the higher tropospheric levels at which they might be transported towards the east, in the direction of Natal, where subsidence would then bring the air to the surface again.

The alternative for transport towards Natal is to participate in the Walker cell circulation motion. The large scale motion of the lower atmosphere in so-called circulation cells is a cause-effect relationship between solar energy input and irregular heating at the surface. The best well-known circulation cells are the Hadley cell causing upward motion of air in the equatorial belt associated with the ITCZ, the Inter-Tropical Convergence Zone, which closes its circuit in a north-south direction, and the Walker cell, also associated with upward motions but over tropical continental areas, closing circuit in an east-west direction. The resulting real circulation is, of course, the result of contributions from both systems. The Hadley cell and the ITCZ are more active northward of the equator in the longitudinal region of the South American continent. The upward motions in the Hadley cell are practically all contained in the troposphere, as shown by Danielson (1979). It is, therefore, unlikely that the descending branches of the Hadley cell can bring higher O_3 values over the Natal area, from a possible exchange with the lower stratosphere.

In a region of high precipitation rates, such as the Amazon, heat is released to the atmosphere through condensation which keeps the ascending motion. In a region of low precipitation rates like the subtropical South Atlantic the air in a given column of air is mostly cooled down by net radiative losses from which descending motions or subsidence results. In this area the flow circuit between ascending and descending motions is closed by eastward flows above about 400 mb and westward flows in the mixing layer.

Upward and downward motions are very difficult to measure because the vertical velocities are very small, of the order of cm/s. It is known, however, that upward motions are associated with low surface pressures and high concentrations of clouds. Downward motions, on the other hand, occur in high surface pressure areas with clear skies. A good correlation can therefore be expected between circulation of the lower atmosphere and precipitation maps.

A precipitation map is shown in Figure 4 for the JJA sessonal period (Nobre, 1983), with basis on data published by Jaeger (1976). Features of interest that can be seen, for example in Figure 4, are the very dry regions in the South Pacific and South Atlantic, on both sides of South America, and more concentrated spots, where large precipitation rates are found, specially close to the center region of the Amazon forest. The slanted lines hatched areas limit regions of precipitation rates of less than 1 mm/day. This general picture of air rising over the Amazon, producing cloudiness and rain, and the descending air east and west of the continent, producing the rather dry high pressure centers, is present in all seasons, the largest difference being the northwest-southeast motion of the center of maximum precipitation. It appears therefore that this rain frequency map can provide a good idea of the atmospheric circulation in the region of our interest. On the basis of rain maps for the four seasonal periods we have produced Figure 5, which shows smoothed precipitation rates for 4 diferent locations of interest. The dry season is found between the JJA and SON periods.

From vertical motion maps, similar to those shown in Figure 6, it can be seen that Natal is within a region of downward motion from June through November, probably in the whole mixing layer. The hatched areas show upward motions. The JJA period in Figure 6a shows subsidence over most of Brazil at the 500 mb level. This is the dry season period during which the vegetation is set on fire. In the following SON period, (Figure 6b), ascending motions of the Walker cell start to dominate again, south of the equator, which could transport lower tropospheric gases to the upper tropospheric levels. However, the Natal area, the whole South Atlantic, and the Southern part of Africa, continue in the subsidence region.

In what follows we suggest a completely different origin for the larger SON O_3 values seen at Natal, that is, the transport of burning products directly from the African continent. The winds in the South Atlantic area have the right direction for the transport of gases to Natal, and large areas of grasslands, savannah-like environments exist in Africa. The extension of areas of low bushes and grassland on the African continent appears to be even larger than the equivalent cerrado region of Central Brazil. As indicated in Figure 7, the African savannah dominates a large area south of 5°S, up to about 20°S, spreading practically over all continent in the east-west direction. The central point of this African savannah land is approximately at 10°S, 20°E indicated by a X in Figure 7. A quick analysis using again the rain maps, as well as the vertical velocities of Figure 6, shows that the high pressure low rain rate area of the South Atlantic extends well into Southern Africa, specially in the JJA period. Figure 5 shows that the dry season in Africa, around coordinates of the savannah region, coincides with the dry period of the Brazilian cerrado. This is before the dry period at Natal and, therefore, enough time is available for transport to Natal. Thus, the contribution of pollution gases from biomass burnings in the African savannah reaching Natal seems a real possibility. Because the burnings should occur during periods of large scale subsidence, the lower troposphere over Africa and the South Atlantic would keep the pollution gases highly concentrated, below the temperature inversion, and the prevailing westward winds would take this air in the direction of Natal. Although the distance from the source is much larger in this case, the displacement of an air parcel between the source and Natal might be shorter, in comparison to the Walker cell mechanism. It should be noted, furthermore, that the dilution factor would be much less in this case because the transport would occur in a region of subsidence in practically the whole transport area.

Wind streamline maps for the area in question differ somewhat from author to author, probably owing to the absence of permanent monitoring stations over the ocean. All authors, however, agree to the fact that the wind streamlines leave the Southern African continent reaching out over the South Atlantic to the north-east coast of Brazil. On the basis of data discussed in Boogaard (1977) and Hastenrath (1985) we show these streamlines in Figure 7. We only show those streamlines that could participate in the transport mechanism we are discussing. It can be seen that in the July season, that is, in the dry period, air from the southern part of the African savannah is in fact directed towards Natal. We plan the calculation of isentropic trajectories for a more quantitative discussion in the near future. It seems, however, that transport in the Walker cell has a much longer distance between source and monitoring station than the other two mechanisms and, therefore, besides transport being of longer duration, the gaseous dilution would be largest in this case.

If we assume a typical vertical velocity of 2 cm/s, in the descending branch, and 10 cm/s in the ascending one, for a height of about 7 km (or higher) for the eastward flow, the vertical displacement of the gases would take at least 5 days, and with a horizontal wind velocity of 5 m/s, and a distance of only 2000 km between ascending and descending branches, another 6 days will elapse, making a total transit time of 11 days in a most favorable condition. The direct transport from Africa would take about 15 days, but in this case the dilution factor may be much smaller, as mentioned before, because the transport is performed in a region of descending air.

In summary, it appears that direct injection of gases from burnings in the cerrado into the upper tropospheric levels through dry convection, or direct transport from the African savannah, are more likely to be able to contribute to contaminated air flow towards Natal than transport through the Walker cell circulation.

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FIGURE CAPTIONS

Fig. 1 - The map of Brazil showing the different kinds of vegetation and the location of ozone monitoring sites.

Fig. 2 - Typical pattern of the diurnal variation of surface ozone.

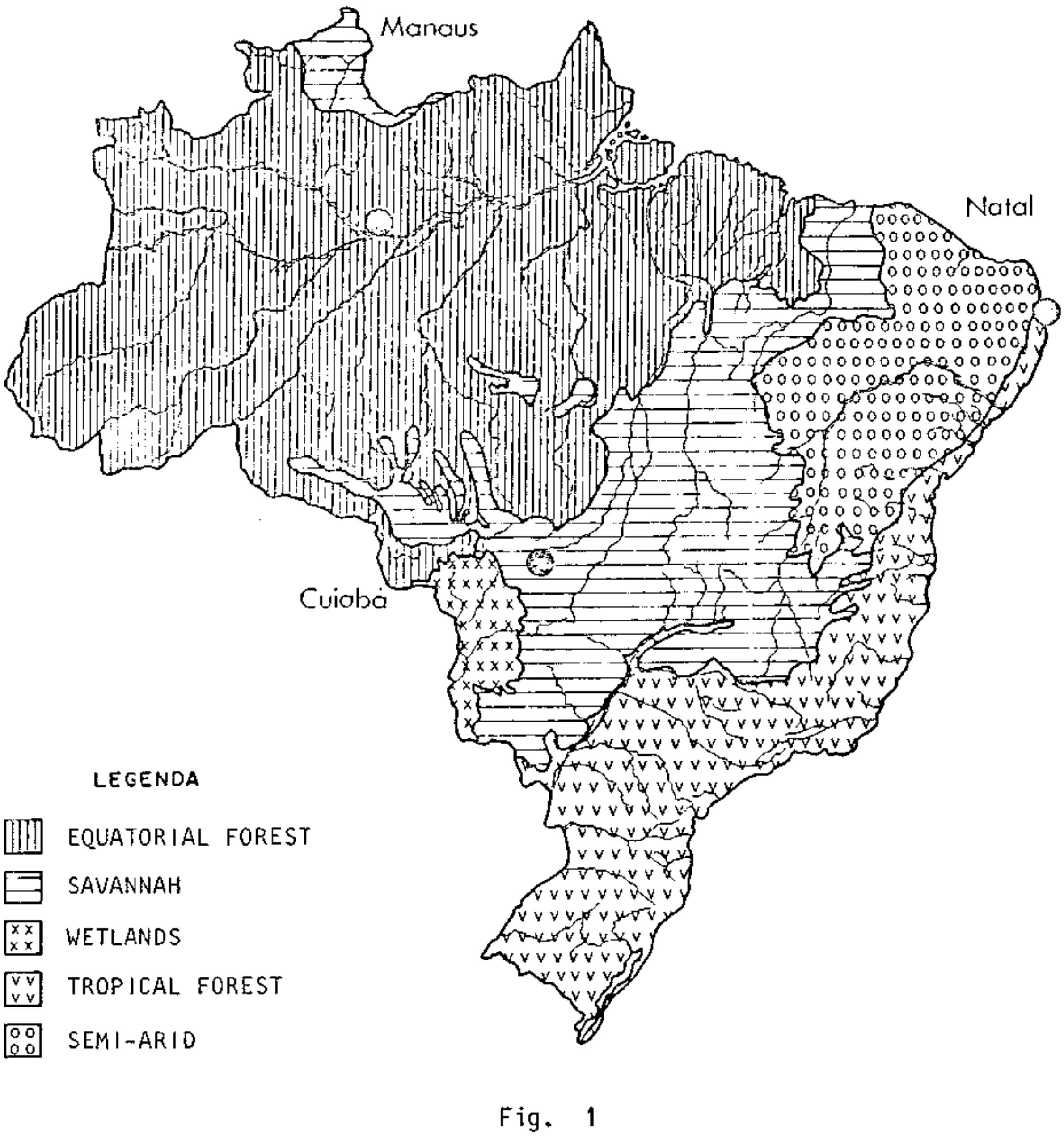
Fig. 3 - Seasonal variation of ozone concentrations at several pressure levels as measured at Natal.

Fig. 4 - Precipitation rate map (mm/day).

Fig. 5 - Mean precipitation rates for Cuiabá, Manaus, Natal, and Africa (10^{o}S, 20^{o}E).

Fig. 6 - Vertical velocity map. Ascending motions are hatched.

Fig. 7 - Wind streamlines that can bring air from Africa to Natal.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LEGENDA
EQUATORIAL FOREST
SAVANNAH
WETLANDS
TROPICAL FOREST
SEMI-ARID

Fig. 1

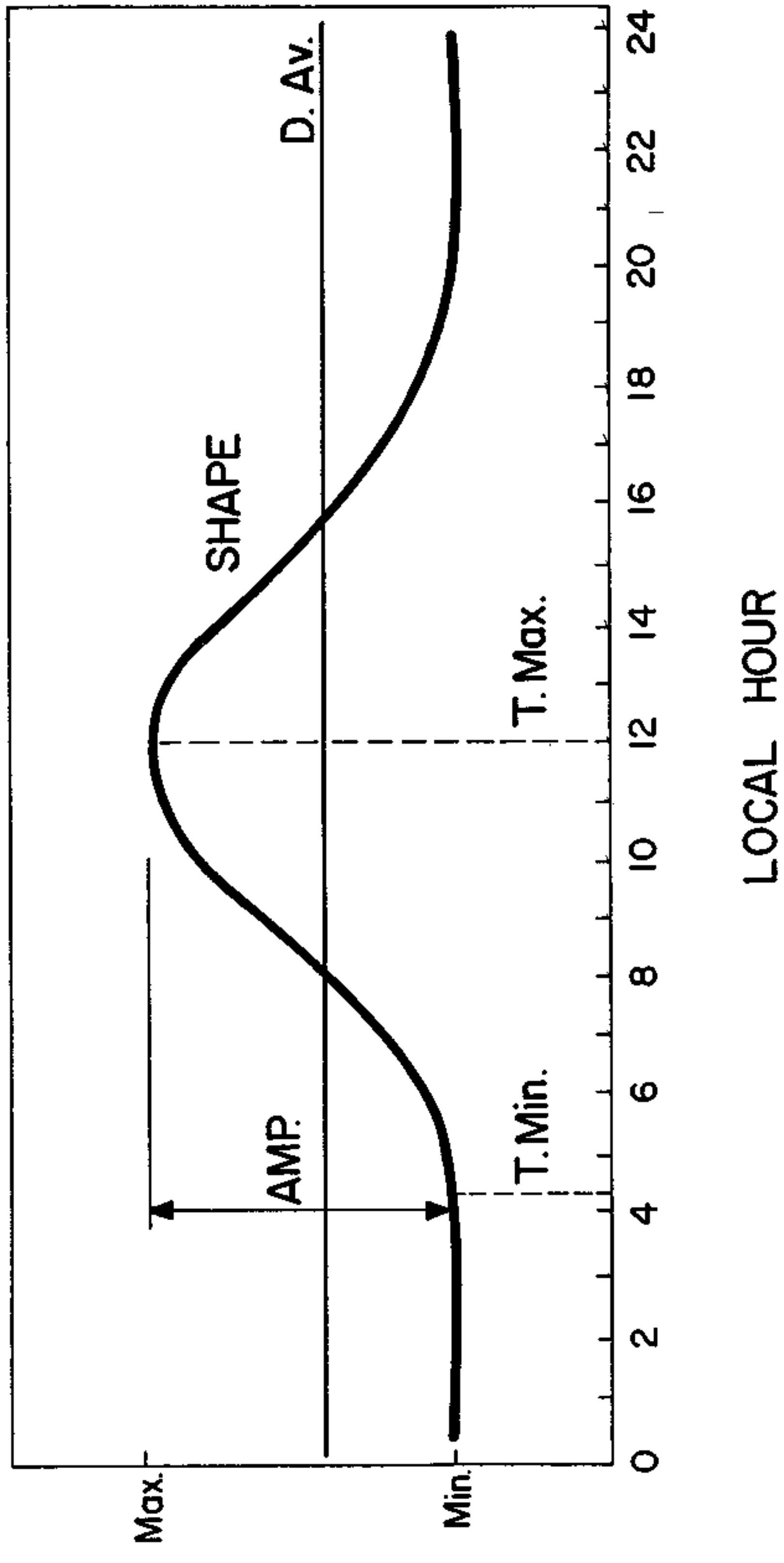
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Fig. 2

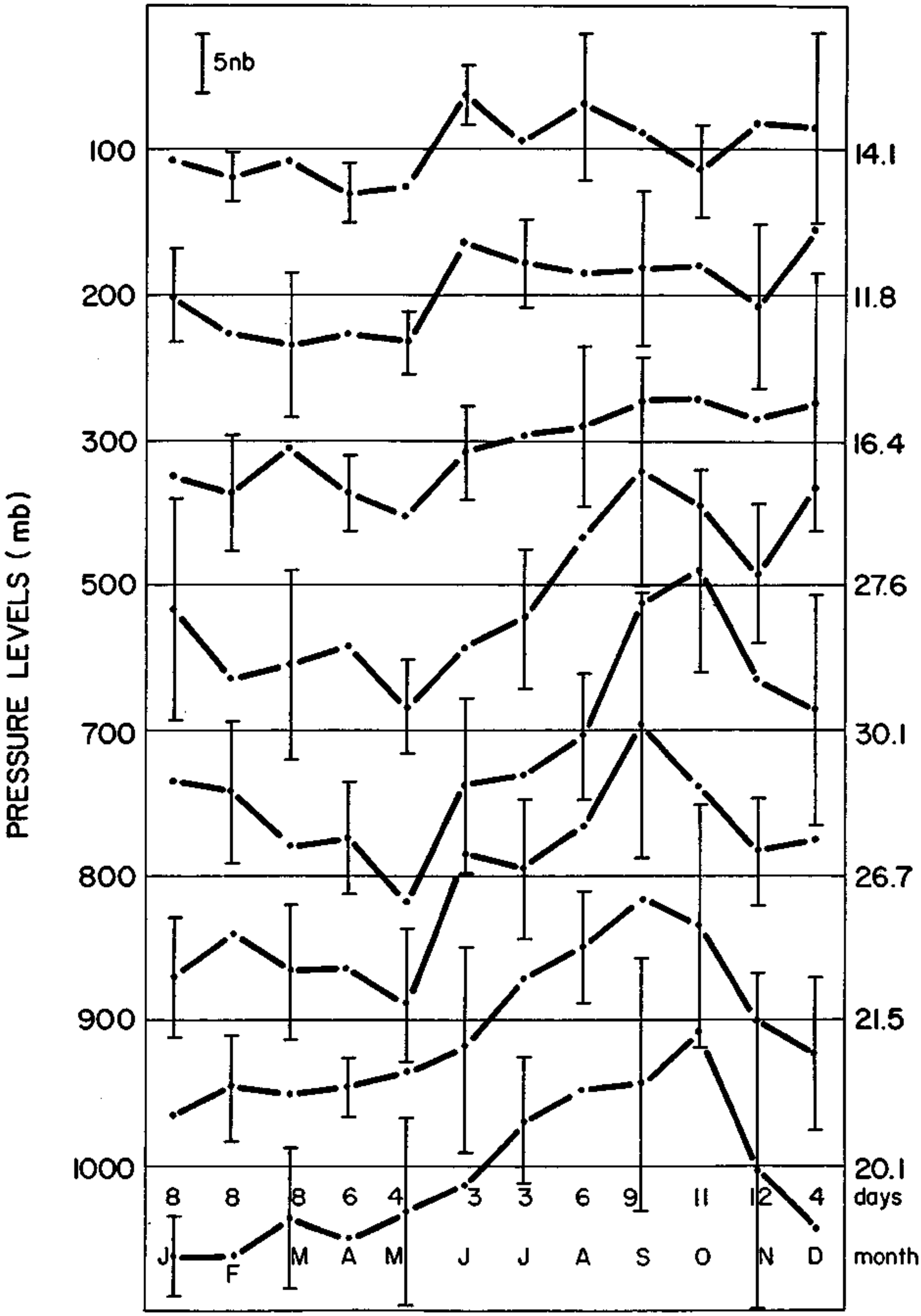
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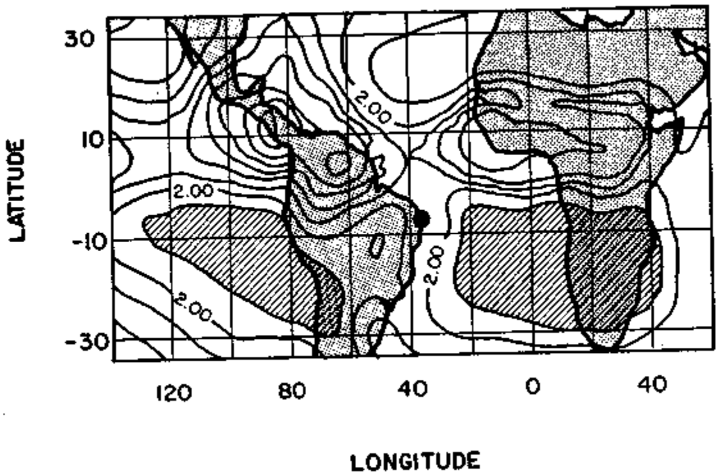
Fig. 3



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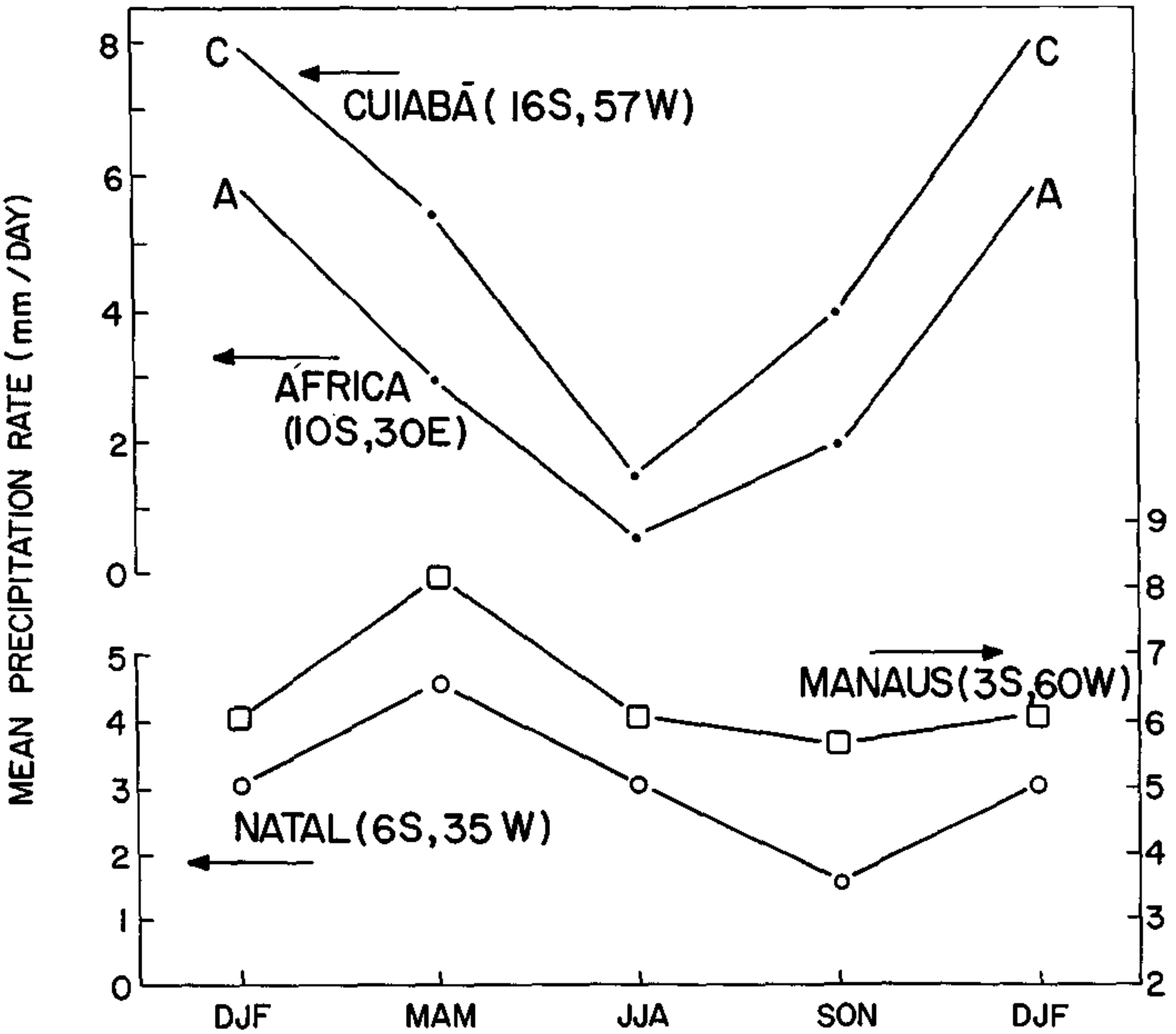
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Fig. 5

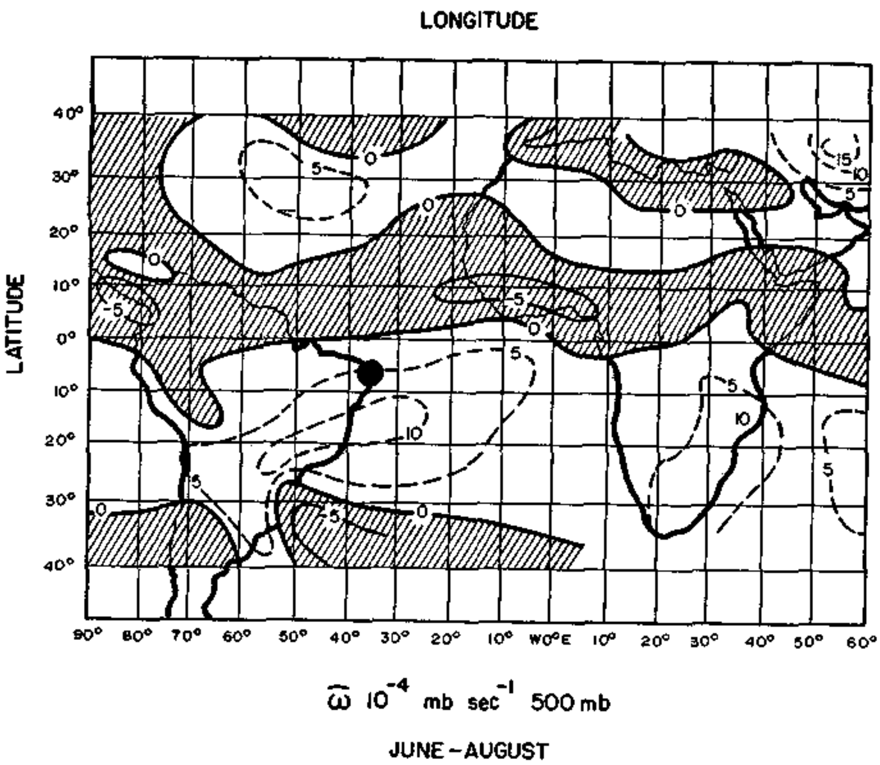
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Fig. 6a

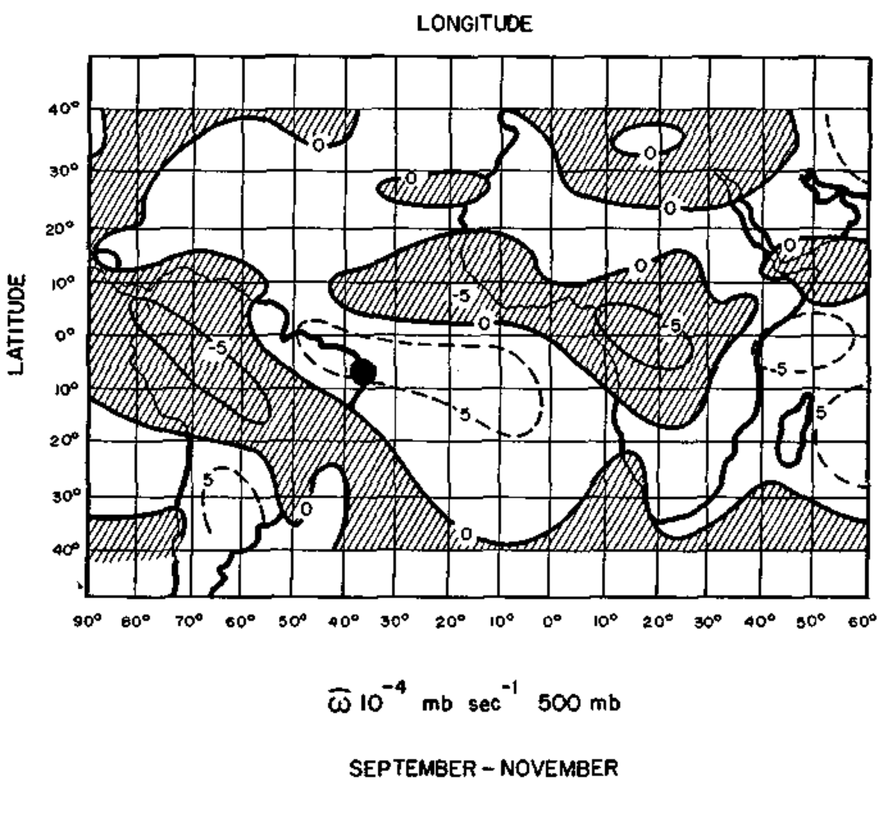
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Fig. 6b

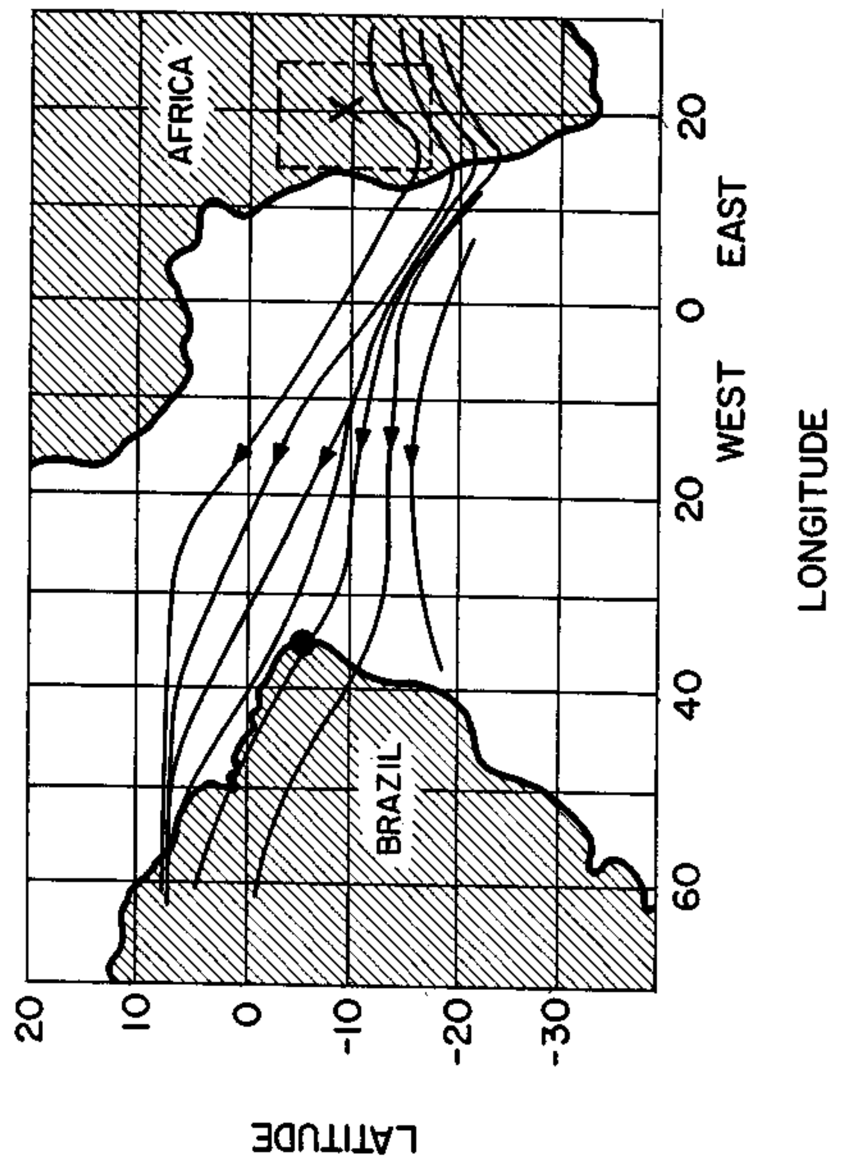
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Fig. 7