

## EXPRESS LETTER

# Constraints on climate forcing by sulphate aerosols from seasonal changes in Earth's spin

Gianluca Sottili

*Istituto di Geologia Ambientale e Geoingegneria IGAG-CNR, Via Salaria km 29,300, I-00015 Monterotondo Stazione, Rome, Italy.*  
E-mail: [gianluca.sottili@uniroma1.it](mailto:gianluca.sottili@uniroma1.it)

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## SUMMARY

Angular momentum exchanges between atmosphere and solid Earth are strongly modulated by variations in global atmospheric circulation. Geodetically determined length-of-day (LOD) fluctuations provide an independent resource to investigate climate changes. Here, I evaluate the effects of volcanic and anthropogenic sulphate aerosols on Earth's rotational energy variations. The period analysed, 1980–2002, shows that the strongest seasonal LOD variations are related to sulphate peak concentrations from the El-Chichón 1982, and Pinatubo and Cerro Hudson 1991 volcanic eruptions. The Earth's rotational energy budget implies that radiative forcing alone cannot produce the observed LOD anomalies. Rather, the required amount of atmospheric kinetic energy can be explained only by a strong influence of sulphate aerosols on energy partitioning into the atmosphere, for example, as sulphate aerosols affect latent heat release and transport during condensation–evaporation–freezing cycles. Overall, the effects of sulphate aerosols on Earth's spin changes are faster than those produced by greenhouse gases.

**Key words:** Earth rotation variations; Atmospheric effects (volcano); Volcanic gases.

## INTRODUCTION

Atmospheric angular momentum (AAM) changes reflect the global-scale dynamic interaction of atmosphere with solid Earth and oceans through a variety of mechanisms involving friction, pressure against topographic barriers and gravity. Specifically, changes in global AAM have been linked to seasonal climate variations (Hide *et al.* 1980; Rosen & Salstein 1985; Salstein & Rosen 1986; Huang *et al.* 2001). Since seasonal AAM variations induce length-of-day (LOD) changes, the Earth's rotation rate may provide a geodetically based way to monitor climate change (Huang *et al.* 2001) on condition that long-term LOD variations are filtered, that is, decadal to secular changes due to geological and astronomical forcing (e.g. Peixoto & Oort 1992).

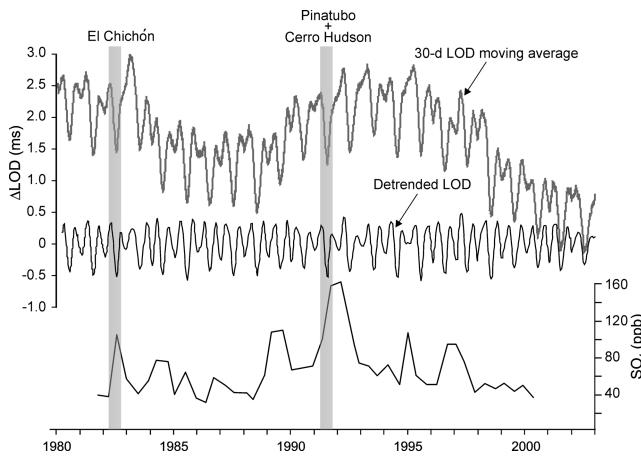
By reflecting solar radiation and acting as cloud condensation nuclei, sulphate aerosols play a key role in the global climate system. Occasionally, volcanoes emit large quantities of sulphur dioxide directly into the atmosphere; the 1991 Pinatubo eruption, for example, released ~30 Tg of SO<sub>2</sub> (Deshler 2008). Sulphate aerosols released during large explosive eruptions spread out globally and reduce the amount of solar energy entering the Earth's atmosphere (Robock 2000) with significant effects on precipitation and atmospheric circulation (Oman *et al.* 2006; Bourassa *et al.* 2012; Timmreck 2012; Iles *et al.* 2013). Recently, a correlation between LOD variations and the occurrence of large eruptions on a global scale (volcanic explosivity index, VEI ≥ 4; e.g. Palladino & Sottili 2012) have

been related to sulphate emissions (Palladino & Sottili 2014). Anthropogenic sulphur exceeds natural sources by a factor of 3 to 4 (35 Tg a<sup>-1</sup> versus 10 Tg a<sup>-1</sup> from 1990 to 2000; Halmer *et al.* 2002) and understanding the effects of sulphate aerosols, from a local to a global scale, represents one of the most important elements of uncertainty in global climate modelling, according to the Intergovernmental Panel on Climate Change (Solomon *et al.* 2007). To reconstruct atmospheric SO<sub>4</sub> concentrations time-series, polar ice caps are considered the best natural records of Earth's atmospheric changes (Cole-Dai *et al.* 1997; Shaheen *et al.* 2013).

Here, the impact of volcanic and anthropogenic atmospheric SO<sub>4</sub> concentrations on angular momentum exchange between atmosphere and solid Earth is evaluated from the energy budget of Earth's spin. Atmospheric sulphate concentrations, derived from polar ice caps (Shaheen *et al.* 2013), appear to be related to statistically significant seasonal LOD anomalies during the analysed 22-yr time period (1980–2002). The analysed time-series also evidences that the strongest seasonal LOD anomalies occurred 1 yr after three large explosive eruptions, that is, El-Chichón 1982, and Pinatubo and Cerro Hudson 1991.

## DATA SET AND ANALYSIS

The high-resolution 1980–2002 record of sulphate aerosols derives from the analysis of a snow pit at the South Pole (Shaheen *et al.* 2013). During the 22-yr period examined here, the yearly



**Figure 1.** LOD changes, expressed as the 30-d moving average, and sulphate aerosols concentrations during the 1980–2002 period. Seasonal LOD oscillation are evidenced by subtracting the annual from the 30-d moving average (detrended LOD curve). The occurrence of El Chichón, Pinatubo and Cerro Hudson volcanic eruptions are also reported. Sulphate concentration data (Shaheen *et al.* 2013) are expressed in ppb.

sulphate concentrations range between 36 to 165 parts per billion (ppb) with two major peaks due to El-Chichón 1982, and Pinatubo and Cerro Hudson 1991 volcanic eruptions, and one unknown event that occurred in 1997 (Shaheen *et al.* 2013). Uncertainty associated with the time of the sulphate peaks is in the order of a few months (Shaheen *et al.* 2013). The LOD data set consists of daily values (Fig. 1) from the International Earth Rotation and Reference System Service (IERS-EOP) database (Bizouard & Gambis 2009), Earth Orientation Center at the Paris Observatory (<http://hpiers.obspm.fr/eop-pc/>). Positive and negative LOD anomalies,  $\Delta LOD$ , are the difference between astronomically measured universal time ( $UT1$ ) and international atomic time (French, Temps Atomique International; TAI) and are given by eq. (1) (Eubanks *et al.* 1985):

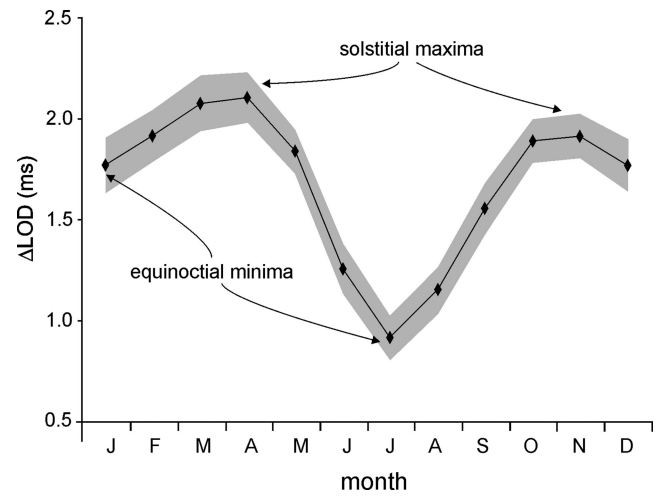
$$\Delta LOD = -LOD_0 \frac{d(UT1 - TAI)}{d(TAI)}, \quad (1)$$

where  $LOD_0$  is the length of a standard day,  $86\,400 \times 10^3$  ms and  $UT1$  is the Universal Time.

The LOD seasonal pattern is analysed as related to possible climatic effects induced by the variability of sulphur gases concentrations into the atmosphere. The choice of LOD seasonal pattern as a proxy to determine changes in global-scale atmospheric circulation is based on evidence for seasonal variations in the atmosphere angular momentum resulting from the opposite contributions by subtropical jets in the Northern and Southern hemispheres (Rosen & Salstein 1985) and from El Niño Southern Oscillation and quasi-biennial oscillation (Chao 1989). Moreover, the subseasonal LOD variations were found to be related primarily to subtropical wind variations, that is, to the Madden–Julian oscillation (MJO), the dominant element of the intraseasonal (30–90 d) variability in the tropical atmosphere (Neef & Matthes 2012).

There are two main components to the analysis: the first is to identify statistically significant features of seasonal LOD responses to atmospheric sulphate aerosols concentrations; secondly, the timescale of significant levels of correlations between  $SO_4$  and LOD changes is examined in order to determine the dynamics of sulphate forcing on LOD.

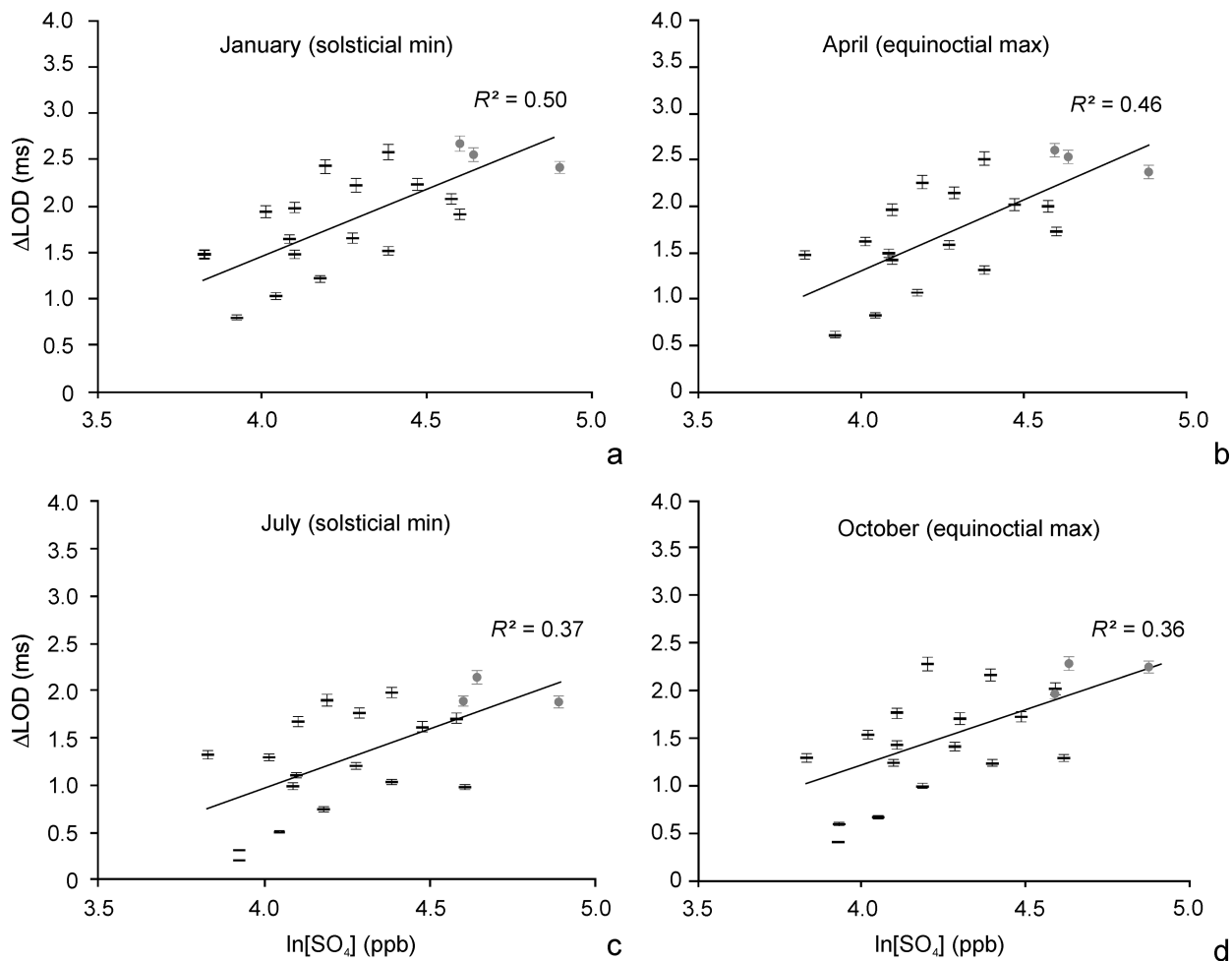
Overall, it appears that the 22-yr LOD pattern is very variable, with total multiyear changes ( $\Delta LOD$ ) up to  $\sim 3$  ms (Fig. 1). In order



**Figure 2.** Monthly  $\Delta LOD$  mean values (1980–2002 period) derived from the detrended LOD curve reported in Fig. 1. The plot evidences two solstitial maxima (in April and in October) and two equinoctial minima (in January and in July).  $\Delta LOD$  values are calculated from the monthly average of daily LOD values (see data source in the text). Grey shading is the year-to-year standard deviations.

to filter the multiyear trend, the 1-yr moving average was removed from the original LOD signal (see detrended  $\Delta LOD$  signal, Fig. 1b). The monthly  $\Delta LOD$  pattern (Fig. 2) shows seasonal, near-sinusoidal oscillations with two annual minima, in January and July, 1 month after solstices, and two maxima, in April and October, 1 month after equinoxes. In the following analysis, to reduce the effects of possible outliers, ‘seasonal  $\Delta LOD$  minima’ are defined as the daily  $\Delta LOD$  values below the 10th percentile recorded in January and in July, respectively, and ‘seasonal  $\Delta LOD$  maxima’ are the daily  $\Delta LOD$  values above 90th percentile in April and in October, respectively. Linear interpolations in Fig. 3 show that, for increasing values of  $SO_4$  concentration, after 1 yr both seasonal  $\Delta LOD$  minima and maxima increase, that is, Earth’s spin decelerates as the sulphate concentrations increases.

The significance of the measured positive relationship between sulphate aerosols concentrations and seasonal LOD changes was tested using the Monte Carlo technique (climate time-series analyses through Monte Carlo technique are also reported in Ting *et al.* 2011 and Iles *et al.* 2013). This meant reshuffling the 22-yr sulphate aerosols time-series like a deck of cards to obtain 1000 new square of regression coefficients,  $R^2$ , between randomized time-series of sulphate aerosols concentrations and changes of seasonal  $\Delta LOD$  minima and maxima. The range of variability of  $R^2$  provides the confidence intervals and only results beyond 2–98 per cent from the Monte Carlo method were considered significant. Highly significant positive relationships ( $>99.5$  per cent confidence) of  $SO_4$  concentration on seasonal LOD changes are observed for the January solstitial minimum ( $R^2 = 0.50$ ) and the April equinoctial maxima ( $R^2 = 0.46$ ). Also, the July solstitial minimum ( $R^2 = 0.37$ ) and the October equinoctial maxima ( $R^2 = 0.36$ ) are positively and significantly related ( $>98$  per cent confidence) to sulphate concentration values. The resulting linear interpolation from the four  $\ln[SO_4]$  versus  $\Delta LOD$  plots reported in Fig. 3 ranges between  $1.05 \text{ ms } \ln[SO_4]^{-1}$  (January) and  $1.57 \text{ ms } \ln[SO_4]^{-1}$  (October). On these grounds, the maximum recorded yearly change of  $SO_4$  concentration (0.8 ln-unit, from 1981 to 1982) would correspond, after 1 yr, to LOD changes between 0.8 to 1.3 ms; these values fit well with the geodetically determined year-to-year seasonal LOD changes, which



**Figure 3.** Changes in length of day ( $\Delta\text{LOD}$ ) 1 yr after  $\text{SO}_4$  concentration measurements (1980–2002 period). During the (a) January solstitial minima and (b) April equinoctial maxima,  $\text{SO}_4$  concentration and seasonal LOD changes show the most significant positive relationship (>99.5 per cent confidence; see text for explanation) with respect to the (c) July solstitial minima and (d) October equinoctial maxima (>98 per cent confidence). Notably, the highest values of sulphate concentrations follow the El Chichón, Pinatubo and Cerro Hudson volcanic eruptions (grey circles in the plot) and correspond to the highest  $\Delta\text{LOD}$  values. Vertical bars are the standard deviations. Sulphate concentrations from Shaheen *et al.* (2013).

vary by +1.2 ms from 1982 January to 1983 January, by +0.8 ms from 1982 April to 1983 April and from 1982 July to 1983 July and by +0.4 ms from 1982 October to 1983 October.

Then, the time response of  $\Delta\text{LOD}$  to sulphate peak concentrations is investigated (Fig. 4). When seasonal changes of  $\Delta\text{LOD}$  minima and maxima are examined across the  $-2$  to  $+3$  yr time window (i.e. relative to year 0, the year of  $\text{SO}_4$  measurements; Fig. 4), a significant degree of positive correlation (>98 per cent confidence) is found since the second half of year 0, while the highest confidence values of correlation are attained in the first half of year  $+1$ . Overall, from seasonal  $\Delta\text{LOD}$  values in Fig. 4, it appears that changes in the Earth's spin associated with a given  $\text{SO}_4$  concentration last for  $\sim 1.5$  yr. When considering the yearly  $\Delta\text{LOD}$  values, that is, annual mean of the daily  $\Delta\text{LOD}$  values, the sulphate concentrations are positively correlated to  $\Delta\text{LOD}$  (significance level >98 per cent) during year  $+1$  (Fig. 4).

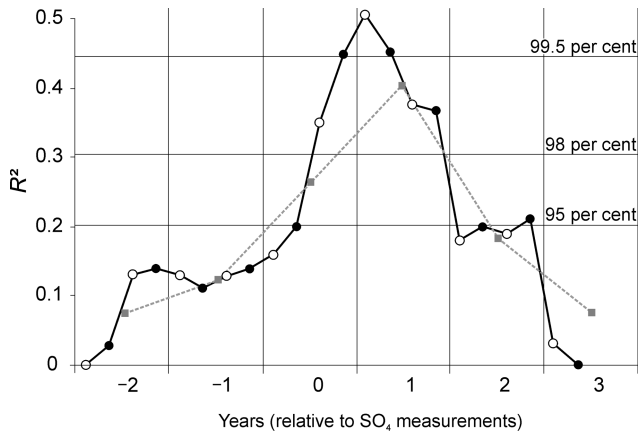
#### EARTH'S SPIN ENERGY BUDGET AND SULPHATE AEROSOLS FORCING

The above analysis shows that, during the 1980–2002 period, changes in the Earth's rotation are positively correlated with

sulphate aerosol concentration recorded at the South Pole. The null hypothesis of a random, positive correlation between  $\Delta\text{LOD}$  and sulphate concentration values can be rejected at the <2 per cent significance level using the Monte Carlo method. The statistically significant level of a positive correlation is higher during boreal winter and spring. The highest sulphate aerosol concentrations recorded during the 22-yr period was originated by large eruptions (i.e. El-Chichón 1982; Pinatubo & Cerro Hudson 1991) and was correlated with the relatively highest seasonal Earth's spin decelerations. Based on aforementioned evidence, it is possible to draw some considerations from the Earth's rotational energy budget. Since the atmosphere does not accelerate in the long-term, the AAM it gains must equal the amount of angular momentum it ultimately returns to solid Earth, even if departures from the average are observed for discrete time windows, and these departures are related to changes in the Earth's spin (e.g. Reid & Gage 1984).

First, the energy variations associated with the observed seasonal LOD changes is considered. The Earth's rotational energy,  $E_T$ , can be derived from

$$E_T = \frac{1}{2} C \omega^2, \quad (2)$$



**Figure 4.** Square of regression coefficients,  $R^2$ , between LOD changes and  $\text{SO}_4$  atmospheric concentrations (1980–2002 period) evaluated within the  $-2$  yr to  $+3$  yr time window with respect to year 0 of  $\text{SO}_4$  measurements. Horizontal lines (95, 98 and 99.5 per cent) refer to confidence limit for  $R^2$  values based on the Monte Carlo technique (see text for explanation). As of the second half of year 0, it appears that solstitial minima (empty circles) and equinoctial maxima (filled black circles) LOD values are significantly and positively correlated with sulphate concentration values, with the highest degree of correlations during the first half of year  $+1$ . Yearly mean  $\Delta\text{LOD}$  values (grey squares; see text for explanation) are significantly correlated with  $\Delta\text{LOD}$  only in year  $+1$ . The duration of the effect of  $\text{SO}_4$  concentrations on seasonal  $\Delta\text{LOD}$  signals lasts  $\sim 1$ – $1.25$  yr.

where  $C = 8.034 \times 10^{37} \text{ kg m}^2$  is the moment of inertia about the polar axis and  $\omega = 7.29246 \text{ rad s}^{-1}$  is the angular speed of the Earth. From eq. (2), the highest sulphate-induced LOD changes (i.e.  $\sim 0.8$ – $1.3 \text{ ms yr}^{-1}$ , see above) during the analysed 22-yr time-series are associated to yearly  $E_T$  variations ranging from  $1 \times 10^{21}$  to  $5 \times 10^{21} \text{ J}$ . If this LOD change was induced entirely by aerosols radiative forcing (i.e. reduction of solar energy influx), this deficit of energy would represent  $\sim 0.1$  per cent of the annual solar irradiance on Earth (i.e.  $\sim 3 \times 10^{24} \text{ J a}^{-1}$ ; Bolton 1977). By way of comparison, the albedo increase by volcanic aerosols after the Pinatubo 1991 eruption was in the same order of magnitude (i.e.  $\sim 0.007$ ; Wielicki *et al.* 2005). Since atmospheric winds (ground to 10 hPa) are the dominant mechanism ( $> 85$  per cent) exciting seasonal LOD variations with a minor contribution from oceanic currents ( $\sim 5$  per cent; Rosen & Salstein 1985; Gross *et al.* 2004), and if we consider that the direct generation and dissipation of atmospheric kinetic energy represents only  $\sim 1$  per cent of mean influx of solar energy on Earth (Peixoto & Oort 1992), it follows that the observed variations of  $E_T$  cannot be explained in terms of sulphate aerosol radiative forcing alone: that is, only a small fraction of the changes in the radiative forcing is available to drive changes in atmosphere kinetic energy. Indeed, the observed  $E_T$  variations must be related to a different partition of the available atmospheric energy. A possible explanation concerns the role of sulphate aerosols on the water evaporation–condensation–freezing cycle (Rosenfeld *et al.* 2008). In fact, the latent heat associated with the water evaporation–condensation–freezing cycle involves  $\sim 23$  per cent of the solar radiation input on Earth (Rosenfeld 2006). Thus, in the context of the balance of the Earth's rotational energy, microphysical aerosol effects (e.g. when acting as cloud condensation nuclei) better satisfy the obtained energy budget constraints, while only a minor role ( $> 2$  order of magnitude smaller in terms of energies involved) should be attributed to radiative forcing.

This is also supported by evidence from climatic and meteorological time-series. In fact, detection and attribution analysis indicates

that precipitation changes on a global scale over the past 50 yr have been forced principally by volcanic aerosols (Gillett *et al.* 2004) with effects on the monsoon system (Oman *et al.* 2006; Timmreck 2012) the heat balance between land and ocean (Joseph & Zeng 2011) and the amount of tropospheric and stratospheric water vapour (Forster & Collins 2004). In this regard, a test of climate feedback by water vapour after the 1991 Pinatubo eruption shows how the observed global temperature response is only reproduced in the global general circulation model (GCM) if water vapour changes are included in the radiative calculations (Soden *et al.* 2002). In fact, the global precipitation anomalies induced by Pinatubo-like eruptions, estimated in the order of a few  $\sim 10^{-2} \text{ mm d}^{-1}$  (Iles *et al.* 2013), correspond to  $\sim 10^{21} \text{ J a}^{-1}$  in terms of latent heat release.

Now, I consider the relative effects of sulphate aerosols versus greenhouse gases on Earth's rotational energy budget. On a multidecadal scale, transient climate change modelling indicates that, to an increase of the present-day atmospheric  $\text{CO}_2$  concentration by a factor 3, would be associated a variation of global (atmosphere + solid Earth) angular momentum,  $\Delta M$ , by  $\sim 4 \times 10^{25} \text{ kg m}^2 \text{ s}^{-1}$  (Huang *et al.* 2001). By way of comparison, the sulphate aerosol forcing of  $\Delta M$ , can be evaluated through the relationship  $\Delta\text{LOD} \cong 0.168\Delta M$  proposed by Peixoto & Oort (1992), where  $\Delta\text{LOD}$  is expressed in milliseconds and  $\Delta M$  is expressed in  $10^{25} \text{ kg m}^2 \text{ s}^{-1}$ . On these grounds, the obtained response of seasonal  $\Delta\text{LOD}$  to sulphate aerosols by  $\sim 1 \text{ ms yr}^{-1}$ , corresponds to a  $\Delta M$  change by  $\sim 0.17 \times 10^{25} \text{ kg m}^2 \text{ s}^{-1}$ , that is,  $> 1$  order of magnitude faster than those produced by long-term greenhouse gases.

## CONCLUDING REMARKS

Sulphate aerosols affect the seasonal angular momentum transfer from atmosphere to solid Earth by altering the energy partition in the atmosphere system. From the above results, it appears that an increase in sulphate aerosols corresponds to an increase in angular momentum transfer from atmosphere to solid Earth. Specifically, since fluctuations of Earth's spin are induced by tangential stresses in turbulent boundary layers and by normal (pressure) stresses acting on irregular topography (Hide *et al.* 1997), the observed deceleration in the Earth's spin must be related to an increase in the kinetic energy of the global atmospheric circulation. The implication of these order-of-magnitude arguments is that the amount of energy involved in the process is too large to derive from aerosol radiative forcing alone. Rather, by involving significant amount of latent heat, the complex interplay between the radiative and microphysical effects of aerosols on the evaporation–condensation–freezing cycle result in global-scale convection and atmospheric circulation changes and, finally, in significant LOD variations. Specifically, the physical mechanism explaining the link between sulphate aerosol concentration increase and Earth's spin deceleration is compatible with the well-known positive relationship between aerosols concentrations, height of the tropical tropopause and global angular momentum of the atmosphere (Reid & Gage 1984). In this perspective, sulphate aerosols, by enhancing the upward heat transport during the evaporation–condensation–freezing cycle (Rosenfeld *et al.* 2008), may induce significant anomalies in the height of the tropical troposphere (e.g. Wu *et al.* 2013) and alter the global angular momentum budget of the atmosphere (e.g. Reid & Gage 1984) that requires an upward transport of angular momentum in the tropics and a downward transport at mid-latitudes.

The results of this study suggest that simple models involving only surface cooling induced by aerosol radiative forcing largely underestimate the effects of aerosols on atmospheric circulation.

Innovative experimentations are needed to model the energy budget of the observed aerosol-induced LOD changes. By quantitatively evaluating the aerosol forcing of Earth's rotational energy budget, the approach proposed here can contribute to our understanding and future ability to model climate changes.

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