Using satellite observations to quantify biomass burning emissions of NO_x and hydrocarbons in the Tropics

New Investigator Program in Earth Science

NAG5-10637

Final Report
(March 2001 - March 2005)

Submitted to:

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1. Introduction

This is the final report for NAG5-10637 "*Using satellite observations to quantify biomass burning emissions of NO_x and hydrocarbons in the Tropics*", funded through the New Investigator Program between March 2001 and March 2005. This period includes a 1-year no-cost extension of the original award.

This report summarizes our accomplishments during the duration of the grant. Section 2 focuses on the research component of this work, while section 3 describes the education component. The personnel supported under this project is given in section 4. Section 5 lists publications resulting from NASA support and section 6 provides a list of conferences and seminars where the results were presented.

2. Summary of Research Accomplishments

2.1. Original Scientific Objectives and Tasks

The tropical troposphere is characterized by large biomass burning emissions, which play a significant role in the global budgets of radiatively important aerosols and gases. However, quantitative estimates of biomass burning emissions of O_3 precursors, in particular nitrogen oxides (NO_x=NO+NO_2) and hydrocarbons, remain highly uncertain. This uncertainty severely limits our ability to assess the present and future effects of tropical biomass burning on tropospheric ozone and climate.

Satellite observations of solar backscatter from the Global Ozone Monitoring Experiment (GOME) offer considerable potential for improved mapping and understanding of biomass burning in the tropics. GOME is an instrument aboard the European ERS-2 satellite, which was launched in April 1995 into a polar sun-synchronous orbit and has been providing continuous nadir spectra of the Earth's atmosphere since then. GOME can measure columns of NO_2, CH_2O (formaldehyde), and ozone with a horizontal resolution of 320 km×40 km, yielding complete coverage of the globe in 3 days.

Our 3-year program of analysis and modeling of data from GOME had two specific objectives: (a) *Determine the factors controlling the regional, seasonal and interannual variability of GOME CH_2O and NO_2 column observations to improve estimates of NO_x and hydrocarbon emissions from biomass burning; and (b) assess the impact of biomass burning emissions of NO_x and hydrocarbon on ozone concentrations above tropical continents.*

2.2. Accomplishments

The research activities supported by NASA as part of this NIP funding have lead to the publication of three peer-reviewed articles and one M.S. thesis (see section 5). In addition, two more papers are expected to be submitted in the coming few months. Below we briefly summarize the results in each of these publications. Our emphasis has broadened considerably compared our original goals. In addition to providing new space-based constraints on biomass burning emissions, we have also examined emissions from fuel combustion and from biogenic sources in soils and vegetation. Furthermore, we have expanded our estimates to the globe instead of only focusing on tropical regions.

2.2.1. "Satellite mapping of rain-induced nitric oxide emissions from soils", L. Jaeglé et al., *J. Geophys. Res.,109*, doi:10.1029/2004JD004787, **2004**.

In this paper, we used space-based observations of NO_2 columns from GOME to map the spatial and seasonal variations of NO_x emissions over Africa during 2000. The GOME observations show not only enhanced tropospheric NO_2 columns from biomass burning during the dry season, but also comparable enhancements from soil emissions during the rainy season over the Sahel. These soil emissions occur in strong pulses lasting 1-3 weeks following the onset of rain, and affect 3 million km^2 of semi-arid sub-saharan savanna.

Surface observations of NO_2 from the IDAF (IGAC/DEBITS/Africa) network over West Africa provide further evidence for a strong role for microbial soil sources. By combining inverse modeling of GOME NO_2 columns with space-based observations of fires, we estimate that soils contribute 3.1±1.7 TgN/yr, similar to the biomass burning source (3.8±2.1 TgN/yr), and thus account for 40% of surface NO_x emissions over Africa. Extrapolating to all the Tropics, we estimate a 7.3 TgN/yr biogenic soil source, which is a factor of two larger compared to model-based inventories, but agrees with observation-based inventories.

2.2.2. "Global partitioning of NO_x sources using satellite observations: Relative roles of fossil fuel combustion, biomass burning and soil emissions", L. Jaeglé et al., *Faraday Discussions*, 130, doi:10.1039/b502128f, **2005**.

We present a simple method to partition global NO_x emissions derived from GOME into fuel combustion, biomass burning and soil emissions for the year 2000. Our resulting *a posteriori* fuel combustion emissions (25.6 TgN/yr) are very close to the GEIA-based *a priori* (25.4 TgN/yr), and uncertainties are reduced by a factor of 2 from ±80% to ±40%. The global *a posteriori* estimate of biomass burning emissions (5.8 TgN/yr) is similar to the *a priori* (5.9 TgN/yr), with a significant decrease in the uncertainty from ±200 to ±80%. *A posteriori* biomass burning emissions over Southeast Asia/India are decreased by 46% relative to *a priori*; but over North Equatorial Africa they are increased by 50%.

We find a 68% increase in soil emissions from 5.3 TgN/yr to 8.9 TgN/yr. The *a posteriori* inventory displays the largest soil emissions over tropical savanna/woodland ecosystems during the wet season, as well as over agricultural regions in mid-latitudes during summer. We estimate that 2.5-4.5 TgN/yr are emitted by N-fertilized soils, at the upper end of previous estimates. Soil and biomass burning emissions account for 22% and 14% of global surface NO_x emissions, respectively. We infer a significant role for soil NO_x emissions at northern mid-latitudes during summer, where they account for nearly half that of the fuel combustion source, a doubling relative to the *a priori*. The contribution of soil emissions to background ozone is thus likely to be underestimated by the current generation of chemical transport models.

This paper was selected as a 'hot article' by the chairman of the Faraday Discussion (http://www.rsc.org/Publishing/Journals/fd/News/FD130Hot.asp), see appendix. The paper was also selected for highlighting in the Royal Society of Chemistry (RSC) 'Chemical Science' supplement (http://www.rsc.org/chemicalscience). The University of Washington issued a press release on our findings, see appendix.

We are in the process of examining the implications of the larger than expected microbial soil emissions on tropospheric ozone production, nitrate deposition and N_2O emissions from soils [*Jaeglé et al.*, 2005b].

2.2.3. "Transport of biomass burning emissions from southern Africa", P. Sinha et al., *J. Geophys. Res.*, 109, doi: 10.1029/2004JD005044, **2004**.

This paper was the result of a collaboration with Ricky Sinha and Peter Hobbs to examine the transport of African biomass burning emissions to the neighboring Atlantic and Indian Oceans during the dry season of 2000. We analyzed observations of CO and ozone from ground-based instruments, ozonesondes and aircraft platforms in and around southern Africa with the GEOS-CHEM model. We quantified the transport of biomass burning emissions through the main two pathways of export: westward to the Atlantic Ocean and eastward to the Indian Ocean. A large fraction of the pollution initially exported westward is eventually transported to the Indian Ocean under anticyclonic conditions. The Indian Ocean is thus the primary net recipient of biomass burning emissions from southern Africa. The model reproduced the long-range transport of biomass burning emissions from southern Africa and South America to Australia, thus explaining observed enhancements of CO over Melbourne in mid-September 2000.

2.2.4. "Using space based observations of NO_2 and HCHO to map biomass burning emissions of NO_x and VOCs over Africa", L. Steinberger, M.S. Thesis, University of Washington, Seattle, **2004**.

In this M.S. thesis, Linda Steinberger examined the spatial and temporal variability of NO_2 and HCHO columns observed by GOME over Africa during the year 2000. The observed columns were compared to simulations from the GEOS-CHEM global model of tropospheric chemistry. We found a high degree of correlation between GOME NO_2 columns and space-based fire indicators from ATSR, VIRS and GBA-2000. In addition, NO_2 and HCHO columns were highly correlated over fire regions, indicating a strong source of VOCs from biomass burning. The model generally captured the levels, as well as the spatial and temporal variability in observed NO_2 columns. In contrast, modeled HCHO columns were too low over biomass burning regions, and did not capture the seasonal variability of HCHO over non-burning regions. This points to a model underestimate of VOC emissions from fires and to an overestimate of isoprene emissions.

In the coming few months, we anticipate submitting a manuscript quantifying biogenic and biomass burning emissions of VOCs over Africa using GOME observations [*Jaeglé et al.*, 2005c].

3. Education Component

3.1. Goals and tasks

The education component focused on developing a web-based interactive teaching tool for atmospheric chemistry, giving students access to recent atmospheric observations and a simple interface to interactively use atmospheric models of radiation, chemistry, and transport

3.2. Accomplishements

3.2.1. Interactive exercises

We have developed five interactive exercises, which are currently being used in the P.I.'s graduate level "Atmospheric Chemistry" class at the University of Washington (taught each year for the past 5 years). Some of these exercises were also used for the undergraduate level class "Climate and Climate Change" for non-science majors. These tools have also been used for teaching and research purposes at other universities and can be viewed at: *http://www.atmos.washington.edu/~jaegle/teaching.html*:

1. **Ozone and UV radiation** -- This module examines the impact of the ozone layer on UV radiation at various levels in the atmosphere. You can change the total amount of ozone and see the resulting effect on the penetration of UV radiation in the atmosphere.
2. **Blackbody radiation --** Calculate the flux distribution of a blackbody as a function of wavelength (Planck function). The user can interactively change the temperature and see the resulting effects on maximum wavelength and total energy energy.

3. **The Sun as a blackbody: Guessing game --** Assuming that the Sun's photosphere behaves as a blackbody, find the temperature of a blackbody that will best match the observed solar radiation spectrum at the top of the atmosphere.
4. **Stratospheric ozone and the Chapman mechanism –** Interactive calculation of the ozone profile in the stratosphere based on the 4 reactions from the Chapman mechanism. You can change the latitude and season of the calculation, and plot the reaction rates.
5. **Global distribution of ozone and its precursors --** Display global concentrations of tropospheric O_3 concentrations and of its precursors (CO, NOx, and related species). The monthly averaged concentrations are calculated with a global three-dimensional model of tropospheric chemistry, GEOS-CHEM.

The interactive web teaching tools are run on a linux web server. The web interface is written in HTML format and the interactive part of the site uses the ION Script software (IDL on the web), which allows to call IDL (Interactive Data Language) procedures to generate plots with user inputs and display the results in text or graphical format.

These web teaching tools were used in several problem sets. These were well received by the students: they enjoyed being able to explore parameter space through the interactive interfaces and thus gain a more intuitive understanding of the concepts behind the problem sets.

In addition, the web interfaces were extensively used during class to illustrate concepts that were discussed. For example the P.I. has used the GEOS-CHEM ozone module (module 5) to contrast the latitudinal and vertical distributions of NO_x, PAN, HNO_3, CO, OH and O_3; illustrate the seasonal variations in OH concentrations; examine the effects of emissions, chemistry and transport in affecting the global distribution of species with various lifetimes; view long-range transport of various pollutants.

3.2.2. Interactive interface to display chemical forecasts

We have adapted the tropospheric ozone module to interactively display forecast results from the GEOS-CHEM model for the NOAA ITCT 2002 (Intercontinental Transport and Chemical Transformation) and the NASA INTEX-A aircraft campaigns. We leveraged funding from NIP with funding from NOAA and NASA to accomplish this, in a collaboration with Daniel Jacob from Harvard and Steven Pawson from GSFC.

The GEOS chemical forecasts were automatically transferred to our server at the University of Washington twice a day. Our interactive web interface was operational for March-May 2002 for ITCT2K2 and June-August 2004 for INTEX-A:

- **ITCT2K2:** *http://marzipan.atmos.washington.edu/cgi-bin/ion-p?page=geos_naps.ion*
- **INTEX-A:** *http://coco.atmos.washington.edu/cgi-bin/ion-p?page=geos_intexa.ion*

Scientists in the field in multiple locations (U.S., Japan, Europe) were able to access the model forecasts with ease and plot the results to design flight plans. The user selects the forecast day, and then chooses the day, species, latitude, longitude, and altitude to plot. Maps of a specific region, vertical sections, curtain plots, vertical profiles and animations could thus be generated interactively based on user-input. The curtain plot generation option of our web site allowed to "fly" the aircraft along way-points in the chemical forecasts so as to optimally design flight plans sampling predicted features.

Since the spring of 2004, our interface has been displaying continuously daily Near-Real-Time GEOS-CHEM global tropospheric chemistry simulations (*http://coco.atmos.washington.edu/cgi-bin/ion-p?page=geos_nrt.ion*). This can be used by ground-based and aircraft missions to rapidly examine the consistency of observations with model calculations. This tool is also used to access model results necessary for satellite retrievals. Current users of the system include Dan Jaffe, University of Washington and Qinbin Li, Jet Propulsion Laboratory.

4. Personnel

The P.I., **Lyatt Jaeglé**, directed all aspects of this project. **Linda Steinberger**, a graduate student, examined the relationships the GOME observations of NO_2 and HCHO and other satellite indicators of biomass burning. Linda graduated with a Master of Science degree in Atmospheric Sciences during the summer of 2004. She was supported by funding from the NIP complemented by matching funds from the University of Washington. Computer and programming support was provided by professional staff in the Department of Atmospheric Sciences.

5. Publications

Peer-Reviewed Publications:

Jaeglé, L., L. Steinberger, R.V. Martin, and K. Chance, Global partitioning of NO_x sources using satellite observations: Relative roles of fossil fuel combustion, biomass burning and soil emissions, *Faraday Discussions*, 130, 10.1039/b502128f, **2005a**.

Jaeglé, L., R.V. Martin, K. Chance, L. Steinberger, T. Kurosu, D.J. Jacob, A.I. Modi, V. Yoboué, L. Sigha-Nkamdjou, and C. Galy-Lacaux, Satellite mapping of rain-induced nitric oxide emissions from soils, *J. Geophys. Res.*, 109 (D21310), doi:10.1029/2004JD004787, **2004**.

Sinha, P., L. Jaeglé, P.V. Hobbs, and Q. Liang, "Transport of biomass burning emissions from southern Africa", *J. Geophys. Res.*, 109(D20), D20204, doi: 10.1029/2004JD005044, **2004**.

Thesis:

Steinberger, L., "Using space based observations of NO_2 and HCHO to map biomass burning emissions of NO_x and VOCs over Africa", *M.S. Thesis*, University of Washington, Seattle, **2004**.

Anticipated:

Jaeglé, L., et al., New constraints on soil NO_x emissions and their effects on ozone, HNO_3 deposition and N_2O emissions, in preparation for *J. Geophys. Res.*, **2005b**.

Jaeglé, L., et al., Space-based observations of the seasonal variations in biomass burning and biogenic emissions of VOCs over Africa during 2000, in preparation for *J. Geophys. Res.*, **2005c**.

6. Presentations of Results at Conferences and Seminars

National and International Conferences:

"Space-based observations of the seasonal variations in biomass burning emissions of NO_x and VOCs over Africa during 2000", L Jaeglé, et al., American Geophysical Union, San Francisco, **December 8-12, 2003**. *Eos Trans. AGU, 84*(46), Fall Meet. Suppl., Abstract A22C-1076. (poster)

"Satellite mapping of rain-induced nitric oxide emissions from soils over Africa", L. Jaeglé, et al., American Geophysical Union, Montréal, Canada, **May 17-22 2004**. *Eos Trans. AGU, 85*(17), Jt. Assem. Suppl., Abstract A31D-03. (oral presentation)

"Global partitioning of NO_x sources using satellite observations: Relative roles of fossil fuel combustion, biomass burning and soil emissions", L. Jaeglé, L. Steinberger, R.V. Martin, and K. Chance, Faraday Discussions 130, Atmospheric Chemistry, Leeds, United Kingdom, **April 11-13 2005**, invited.

Seminars at the University of Washington and other institutions:

"Space-based observations of the seasonal variations in biomass burning emissions of NOx and VOCs over Africa during 2000", L Jaeglé, et al., University of Washington, Department of Atmospheric Sciences, Climate and Remote Sensing seminar, **November 7, 2003**.

"Satellite mapping of rain-induced nitric oxide emissions from soils over Africa", L. Jaeglé, University of Washington, Department of Atmospheric Sciences, **May 27 2004**, invited.

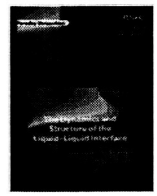
"Using space based observations of NO_2 and HCHO to map biomass burning emissions of NO_x and VOCs over Africa", Linda Steinberger, University of Washington, M.S. Thesis defense, June 2004.

"Partitioning of nitrogen oxides sources using satellite observations", L. Jaeglé, Program on Climate Change seminar, University of Washington, **November 2004**.

"Global partitioning of NO_x emissions using satellite observations", L. Jaeglé, GEOS-CHEM 2nd Users' Meeting, Harvard University, Cambridge, **April 4-6 2005**, invited.

RSC Publishing

Faraday Discussions



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Faraday Discussion 130: Atmospheric Chemistry

Two of the HOT articles presented at the recent Faraday Discussion on Atmospheric Chemistry are available to download, free of charge, on this page.

Modelling NOx sources from space

Global partitioning of NO_x sources using satellite observations: Relative roles of fossil fuel combustion, biomass burning and soil emissions
Lyatt Jaeglé, Linda Steinberger, Randall V. Martin and Kelly Chance, *Faraday Discuss.*, 2005
DOI: 10.1039/b502128f

Modelling NOx sources using satellite observations

Satellite observations have been used to provide important new information about emissions of nitrogen oxides.

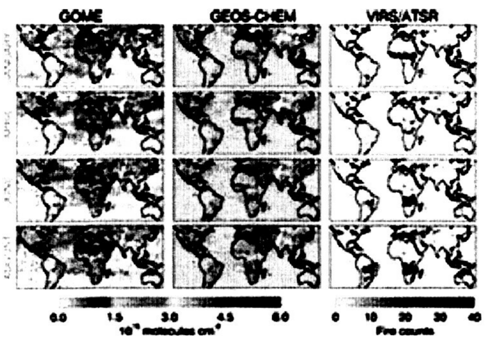
Nitrogen oxides (NOx) are significant in atmospheric chemistry, having a role in ozone air pollution, acid deposition and climate change. We know that human activities have led to a three- to six-fold increase in NOx emissions since pre-industrial times, and that there are three main surface sources of NOx: fuel combustion, large-scale fires, and microbial soil processes. How each of these sources contributes to the total NOx emissions is subject to some doubt, however.

The problem is that current NOx emission inventories rely on 'bottom-up' approaches, compiling large quantities of statistical information from diverse sources such as fuel and land use, agricultural data, and estimates of burned areas. This results in inherently large uncertainties.

To overcome this, Lyatt Jaeglé and colleagues from the University of Washington, USA, used new satellite observations from the Global Ozone Monitoring Experiment (GOME) instrument. As the spatial and seasonal distribution of each of the sources of NOx can be clearly mapped from space, the team could provide independent 'top-down' constraints on the individual strengths of NOx sources, and thus help resolve discrepancies in existing inventories.

Jaeglé's analysis of the satellite observations, presented at the recent Faraday Discussion on "Atmospheric Chemistry", shows that fuel combustion dominates emissions at northern mid-latitudes, while fires are a significant source in the Tropics. Additionally, she discovered a larger than expected role for soil emissions, especially over agricultural regions with heavy fertilizer use.

Turning to space to model atmospheric changes opens up unprecedented possibilities for improving our understanding of the factors controlling the variability in soil and fire emissions, allowing scientists to begin to predict the shifts in these sources in the face of increasing land-use and climate change. Jaeglé is sure of where future challenges lie, commenting, "Over the last few years, new satellite instruments probing the composition of our lower atmosphere have been revolutionizing atmospheric chemistry. The challenge is to use these new global observations to understand and predict the effects of human activities on atmospheric composition, and thus on human health and ecosystems".



Satellite observations of NOx columns from GOME

Distribution of organic compounds during the August 2003 ozone episode

Modelling the ambient distribution of organic compounds during the August 2003 ozone episode in the southern UK
Steven R. Utembe, Michael E. Jenkin, Richard G. Derwent, Alastair C. Lewis, James R. Hopkins and Jacqueline F. Hamilton, *Faraday Discuss.*, 2005
DOI: 10.1039/b417403h

1. Could you explain the significance of your article to the non-specialist?

Many hundreds of different organic compounds are emitted into the atmosphere from human-influenced and natural sources. Their oxidation potentially has harmful impacts on human health and the environment, through formation of secondary pollutants such as ozone and oxidised organic particles. The oxidation mechanisms involve the formation of thousands of intermediate oxidised organic compounds. In this work, a highly detailed representation of the oxidation chemistry has been used to predict the structures and distributions of the oxidised products during the heatwave period in August 2003, and to test their significance in influencing the rates of oxidation and the formation of ozone.

2. What has motivated you to conduct this work?

The chemical mechanism used in this work (the Master Chemical Mechanism, MCM) aims to represent current understanding of the elementary processes involved in atmospheric organic oxidation, based on the results of laboratory studies of reaction kinetics and mechanisms. The MCM is the most comprehensive description of atmospheric organic oxidation available. It can therefore assist in the prediction of the identities and likely ambient concentrations of a large number of organic species. This, in turn, can assist in the development of ambient measurement techniques and the identification of detected, but unknown species.

Jun. 6, 2005 | Science and Tech

Study uncovers dirty little secret: Soil emissions are much-bigger-than-expected component of air pollution

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Nitrogen oxides produced by huge fires and fossil fuel combustion are a major component of air pollution. They are the primary ingredients in ground-level ozone, a pollutant harmful to human health and vegetation.

But new research led by a University of Washington atmospheric scientist shows that, in some regions, nitrogen oxides emitted by the soil are much greater than expected and could play a substantially larger role in seasonal air pollution than previously believed.

Nitrogen oxide emissions total more than 40 million metric tons worldwide each year, with 64 percent coming from fossil fuel combustion, 14 percent from burning and a surprising 22 percent from soil, said Lyatt Jaeglé, a UW assistant professor of atmospheric sciences. The new research shows that the component from soil is about 70 percent greater than scientists expected.

Instead of relying on scattered ground-based measurements of burning and combustion and then extrapolating a global total for nitrogen oxide emissions, the new work used actual observations recorded in 2000 by the Global Ozone Monitoring Experiment aboard the European Space Agency's European Remote Sensing 2 satellite.

Nitrogen oxide emissions from fossil fuel combustion are most closely linked to major population centers and show up in the satellite's ozone-monitoring measurements of nitrogen dioxide, part of the nitrogen oxides family. Other satellite instruments can detect large fires and the resulting emissions also can be measured by the ozone-monitoring experiment, Jaeglé said.

But the satellite also picks up other nitrogen oxide signals not attributable to fuel combustion or burning, and those emissions must come from soil, Jaeglé said.

"We were really amazed that we could see it from space, but because the pulse is so big the satellite can see it," she said.

Soil emissions are seen primarily in equatorial Africa at the beginning of the rainy season, especially in a region called the Sahel, and in the mid-latitudes of the Northern Hemisphere during summer. When the rains come to the Sahel after a six-month dry season, dormant soil bacteria reawaken and begin processing nitrogen. The satellite then detects a sudden pulse of nitrogen oxides, Jaeglé said. Similarly, emissions in the mid-latitudes of the Northern Hemisphere spike during the growing season, spurred by warmer temperatures after a cold winter, but also perhaps magnified by fertilizer use.

"The soil emissions were much larger than we expected," she said. "The biggest areas were the dry topical regions like the Sahel, and in the mid-latitude regions where there is a lot of agriculture."

During summer in North America, Europe and Asia, nitrogen oxides emitted from soil can reach half the emissions from fossil fuel combustion.

"And this is at a time when there are already problems with air pollution," Jaeglé said.

Nitrogen oxides comprise a group of highly reactive gases containing nitrogen and oxygen in varying amounts. Besides producing ozone smog, they help form the dirty brown clouds that often hang over major cities, they contribute to acid rain and they play a role in global climate change.

In addition to equatorial Africa, hot spots for soil emissions include the central plains of the United States; southwestern Europe, primarily the Iberian Peninsula; much of India; and the northern plains of Asia, she said. All of those areas are highly agricultural.

The new research was published in May in Faraday Discussions, a journal of England's Royal Society of Chemistry. Co-authors are Linda Steinberger of the UW; Randall Martin of Dalhousie University in Halifax, Nova Scotia; and Kelly Chance of the Harvard-Smithsonian Center for Astrophysics in Cambridge, Mass. The work was funded by the National Aeronautics and Space Administration's New Investigator Program in Earth Science.

Jaeglé noted that agricultural activity is likely to increase in the future, bringing more fertilizer use. As a result, there could also be even greater soil emissions of nitrogen oxides.

"We don't know how emissions will change, but we now have a way to monitor them from space," she said.

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