Determining relationships and mechanisms between tropospheric ozone column concentrations and tropical biomass burning in Thailand and its surrounding regions

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Abstract

This study aims to determine the variability and trends of tropical biomass burning, tropospheric ozone levels from 2005–2012 in Thailand and the ozone transport from the surrounding regions. Intense biomass burning and tropospheric ozone in this area have a seasonal variability with the maximum generally occurring during the dry season. The northern part of Thailand was observed to have high tropospheric ozone during the dry peak season in April. Forward trajectory analysis determined that ozone sources due to biomass burning in the northern and western surrounding regions (Myanmar, Laos and India) enhance the tropospheric ozone column in northern Thailand. Seasonal variations were also seen for the middle and northeastern regions of Thailand. During August, most biomass burning occurs in Indonesia and Malaysia. However, forward trajectory analysis showed that the effect in the tropospheric ozone column level in the southern part of Thailand is minimal from these regions. Eight-year trends of tropospheric ozone column were also calculated for the different regions of Thailand. However, statistical analysis showed that these trends were not significant. The interannual variability of the tropospheric ozone column concentrations due to El Niño Southern Oscillation were also investigated. It was observed that the best correlation of the tropospheric ozone column with the Oceanic Niño Index (ONI) occurred when ONI was advanced 3 months for the north, northeast and south regions of Thailand and 4 months for the middle region of Thailand.

1. Introduction

Tropospheric ozone (O₃) is a short-lived trace gas that is formed in situ by reactive gases and solar radiation or that is transported from the stratosphere. In the tropics, tropospheric ozone sources due to stratospheric intrusion is excluded (Homeyer et al 2011) by the nature of the Brewer Dobson circulation. However, stratospheric air can also influence ozone levels at high altitudes by lofting air masses over the Himalayas (Dupont et al 2012). At the surface, tropospheric ozone causes health effects to both animals and plants (referred to as bad ozone or smog). Tropospheric ozone has an average atmospheric lifetime of a few weeks (IPCC 2013). This results in a highly variable global distribution that depends by season, altitude and location. Ground-based O₃ is created by the photochemical and chemical reactions between oxides of carbon monoxide (CO), nitrogen (NOₓ), and volatile organic compounds (VOCs) (e.g. Finlayson-Pitts and Pitts 1997, Reid et al 2005, Lamarque et al 2010, Sahu 2012) wherein the cellulosic material and aromatic compounds from open biomass burning are one of the causes of the production. Anthropogenic activities, such as biomass burning are one of the main sources of this bad ozone. Tropospheric ozone is also an important greenhouse gas. Its ambient concentration has increased due to increases in precursor trace gas emissions (e.g. CO, NOₓ, VOCs) from anthropogenic activities. Differences in tropospheric ozone burden have been observed in numerous emissions data mainly due to biomass burning (Lamarque
et al 2010, Cionni et al 2011, Young et al 2013). Jaffe and Wigder (2012) presented a critical review of O₃ production from wild fires focusing on its influence on O₃ production. According to this study, wild fires produced 170 Tg per year of O₃ (3.5%) globally. The majority of O₃ production was high in tropical and equatorial biomes and lower in the other regions.

In Cooper et al (2010), through a compilation of mid-tropospheric ozone observations, indicated that western North America (25°–55°N, 130°–90°W, 3–8 km) experienced a recent (1995–2008) increase in ozone most likely associated with Asian emissions. Amnuaylojaroen et al (2014) applied the different anthropogenic emission inventories in WRF-Chem simulations to examine the O₃ surface mixing ratios for Southeast Asia in March and December 2008. They found that anthropogenic emissions without biomass burning produced only a slight variability of O₃. However, including biomass burning emissions in the simulations increased O₃ variability by about 30%.

The relation of tropical biomass burning and tropospheric ozone had been studied during intense fires over Indonesia in 1996–1998 by Thompson et al (2001). The Indonesian fires of 2006 were studied by Ott et al (2010), Zhang et al (2011), Srivastava and Sheel (2013) and Sahu et al (2014). Results showed that biomass burning emissions are responsible for an increase in O₃. The aerosol and tropospheric ozone are not always in phase due to the maritime continent and intercontinental transport over Indonesia. Aerosol heating also plays an important role in the transport of biomass burning pollution along the vertical by increasing the pollution in the upper troposphere lower stratosphere. El Niño in 2006 was also related to biomass burning and dynamical changes in O₃ concentration.

Liu et al (2002) utilized the GEOS-CHEM chemical transport model driven by assimilated meteorological observations to analyze the extensive observations available from Hong Kong in 1996. The study found that Asian biomass burning makes a major contribution to ozone at <32°N in spring. Chan et al (2003) observed ozone enhancement in spring 1993–1999 over Hong Kong due to photochemistry and transport of ozone precursor-rich air originating from biomass burning emissions from the Indo-Burma region containing Burma, Laos and northern Thailand, and the Indian-Nepal region containing northern India and Nepal as the two most active regions of biomass burning emissions in the Southeast Asia subcontinent. The accompanying trace gas measurements suggest that the O₃-rich air masses contain CH₂Cl₂, a biomass burning tracer and not the general urban emission tracers. The O₃-rich air masses are transported by a westerly wind. The fire hot spots in south China, Philippine islands, Malaysian Peninsula and Indonesian regions were comparatively fewer. However, the maximum fires in this year occurred in Indonesia and, to a lesser extent, Malaysian regions as a result of the strong El Niño phenomenon in that year.

Chan et al (2006) measured tropospheric ozone, carbon monoxide, total reactive nitrogen (NOₓ) and aerosols (PM2.5 and PM10) on the southeastern Tibetan Plateau at Tengchong in Southwest China in the spring of 2004. Fire maps derived from satellite data and backward air trajectories were used to trace the source regions and transport pathways of pollution. The increase of pollutants in the lower troposphere was caused by regional build-up and transport of pollution from active fire regions of the Southeast Asia subcontinent and from northern South Asia which have stronger impacts than that from Central and South China on the abundance of tropospheric ozone, trace gases and aerosols in the background atmosphere of the Tibetan Plateau of Southwest China.

Pochanart et al (2001) measured continuously the mixing ratios of surface ozone at two sites in Thailand. Identical seasonal variations of ozone with dry season maximum and a wet season minimum with a large seasonal amplitude was observed at both sites during 1996–1998. Based on trajectory analysis of ozone data, the long-range transport of ozone under Asian monsoon regime could primarily explain the low ozone mixing ratios in clean marine air mass from the Indian Ocean during the wet season but only partly explain the relatively low ozone mixing ratios in continental air mass from northeast Asia either in wet or dry season. The highest ozone mixing ratios are found in air masses transported within Southeast Asia. The high ozone mixing ratios during the dry season are suggested to be significantly due to the local/sub-regional scale ozone production triggered by biomass burning in Southeast Asia rather than long-range transport effect. The air mass transport during the dry season of 2005–2009 in Chiangmai province in northern Thailand, has been studied by Chantara et al 2012 using 3-day backward trajectories from the HYSPLIT model and using statistical analysis. The results showed that the air mass was transported from three sources: from the Indian Ocean, entering inland to Myanmar; from China, passing through the northeast direction; and from local sources. Stratospheric air also influences the Thailand plume after it is lofted to high altitudes via the Himalayas (Dupont et al 2012). The variation in the tropospheric O₃ mixing ratio over Bangkok, Thailand had been studied by Sahu et al (2013). The study showed that O₃ in the planetary boundary layer had seasonal and diurnal variations. Backtrajectory and fire count data also indicated that the seasonal variations in trace gases were caused mainly by the regional shift in long-range transport and biomass-burning patterns which was affected by the El Niño led extensive fires in Indonesia during 2006.

Duncan et al (2003), Real et al (2007) and Jaffe and Wigder (2012) reported that the wildfire plume, clouds and aerosols can also affect the photolysis rate
by either increasing or decreasing the O3 production due to reduced transmitted solar radiation and photochemical reactions.

In this paper, the following were investigated: (1) the relationship of fire hotspot data and the El Niño Southern Oscillation (ENSO) phenomenon to tropospheric ozone column concentrations over Thailand due to biomass burning and its surrounding regions; and (2) potential source regions of tropospheric ozone columns in Thailand and in the surrounding countries. Fire hotspot data were obtained from Moderate Resolution Imaging Spectroradiometer (MODIS) on-board the Terra and Aqua satellites and the tropospheric ozone column concentration was retrieved from OMI and MLS instruments deployed on the Aura satellite. Fire count density was also calculated to determine the locations where transport simulations will be carried out. The Stochastic Time-Inverted Lagrangian Transport (STILT) model (no chemistry considered) was utilized for the transport simulations (Gerbig et al 2003a, 2003b, Lin et al 2003). Section 2 of the paper describes the methodology in detail. Section 3 shows results and discusses its relationship to the ENSO and transport. Last section outlines the conclusions and recommendations.

2. Methodology

2.1. Study area

In order to fulfill the above-mentioned objectives, Thailand was divided into four regions (solid lines in figure 1) based on land use and human activities. The northern part, as shown in the solid blue box in figure 1, is mostly covered by evergreen forest in the mountainous area. It is surrounded by Laos and Myanmar. The northeast region, as shown in the solid orange box in figure 1, is mostly plateau area and covered by rice cropland and other vegetation. The central region, which is shown in the red solid box in figure 1, is comprised of mixed deciduous forest and plantation area mostly devoted to growing rice. These three regions are warm and rainy during June–September, dry and cool during October–February and dry and hot during March–May. Biomass burning mostly occurs after harvest and forest-clearing during the dry season between December and May. Smog is always observed in these regions during post-harvest in the dry season (December–April), especially in the northern area. In the southern region, the green solid box in figure 1 is bounded at the east and west by oceans and it is in close proximity to Malaysia and Indonesia. The local climate is always hot and humid. There is only few post-harvested product or peat in this region. To study the neighboring influence, the surrounding regions were divided into three regions (dashed boxes in figure 1).

2.2. Fire hotspot data

The MODIS fire hotspot data was used for this study. Two MODIS instruments are on-board the Terra and Aqua satellites, which crosses the equator from north to south in the morning and from south to north in the afternoon, respectively. This results in a global coverage of every 1 to 2 days. Hotspot detection from the MODIS Adaptive Processing System version 5.1 and from the Land Atmosphere Near Real-Time Capability for EOS (LANCE) Rapid Response version 5.0 was utilized in the study. The active fire hotspot products
were systematically developed to consider the global spatial and temporal coverage in the center of 1 km pixel locations to be monitored and flagged for fire detection (https://earthdata.nasa.gov/data/near-real-time-data/). To reduce the potential active fire commission errors, only the high confidence class (80% and above) was used in this study to determine the hotspot data (based on the recommendation of the MODIS user guide). However, clouds and aerosols produce lower fire counts compared to the actual number. The descriptions of the product can be found in the MODIS Collection 5 Active Fire Product User’s Guide Version 2.5 (Geglio 2013).

2.3. Tropospheric ozone column concentrations
The tropospheric ozone column concentration data were collected from the OMI and MLS instruments on-board the Aura satellite. The data was determined by subtracting the MLS stratospheric ozone column from the OMI total column ozone and adjusted for intercalibration using the convective-cloud differential method. The monthly data is provided with a resolution of 1° latitude by 1.25° longitude from October 2004 to present and available for free download via the website http://acdb-ext.gsfc.nasa.gov/Data_services/. The tropical OMI/MLS tropospheric ozone column has been validated with the Southern Hemisphere ADditional OZonesondes (SHADOZ) and shown only small variability. Further details for the OMI/MLS tropospheric ozone retrieval method and validation are discussed by Ziemke et al (2006).

2.4. The Stochastic time-STILT model
The STILT model (without chemistry), driven by the meteorological fields of the Global Data Assimilation System model (1 × 1 degree), was utilized in this study to determine regions where tropospheric ozone is transported from the burning area. This is performed by releasing hypothetical particles forward in time for 3 days from locations having the maximum fire count density. Details of the model can be found in the studies of Gerbig et al (2003a, 2003b) and Lin et al (2003).

2.5. The oceanic Niño index (ONI)
The ONI depicts monthly sea surface temperatures (SST) anomalies across the equatorial Pacific in the Niño 3.4 region (5°N–5°S, 120°–170°W). ONI is a standard used for identifying El Niño (warm) and La Niña (cool) events in the tropical Pacific (Peters et al 2001). The values are derived by using a 3-month running averages of the monthly SST based on 30-year periods and updated every 5 years. The Extended Reconstruckted Sea Surface Temperature v3 was utilized in this study (http://ggweather.com/enso/oni.htm). The description of the product can be found in Xue et al (2003) and Smith et al (2008).

3. Results and discussion
3.1. Relationships to fire occurrence and ENSO
The time series of the ONI and the fire occurrence for each region in Thailand and including the surrounding regions are shown in the top and middle panels of figure 2, respectively. Fire occurrence is a dimensionless quantity that is calculated as the ratio of the
number of fire hotspot observations to the total number of MODIS measurements. The monthly averages of tropospheric ozone column levels for each region in Thailand are shown in the bottom panel of figure 2.

Both the ozone levels and the fire occurrences have variation with the season for every region. Background tropospheric ozone sources come from common human activities (e.g. NOx vehicle emissions), but the tropospheric ozone variation corresponds to the variation of biomass burning. High tropospheric ozone levels were observed during the dry season from February to May of every year, particularly in the north, north east and middle regions of Thailand with variations of 20–40 DU. This corresponds to the more frequent fire occurrences that occurred in those regions. Low tropospheric ozone (10–30 DU) and fire occurrence occur during the wet season from June to July in the southern region.

Eight years of tropospheric ozone data over Thailand show an increasing trend for the northern part of Thailand (of about 0.069 DU/year) and a decreasing trend in other regions i.e. −0.109, −0.255, −0.131, and −0.303 DU/year for north, northeast, middle and southern parts of Thailand, respectively. The t-test of the linear regression analysis during the burning season (February–April) during 8 years shown in table 1 showed that the tropospheric ozone levels are decreasing for all regions but they were not significant in the 95% confidence interval (p-value > 0.05 and |t| < t_{crit} of 2.4469). The highest value of the mean tropospheric ozone level during the burning season is 33.77 DU in the north and the lowest is 26.24 DU in the southern region. However, tropospheric ozone concentrations for the overall 8 years and for the 8 year-burning season months do not show significant trends (at α = 0.05) of tropospheric ozone levels but the seasonal variation of the ozone can obviously be seen in the figure 2.

Biomass burning in the northern part of Thailand area is highest compared to other regions. Including neighboring regions, the fire occurrences were high in the both north and south of Thailand but not much for the middle part of Thailand (red dashed line in the middle panel of figure 2). The high frequency of fire in the north (blue dashed line in the middle panel of figure 2) occurs during February to April and in the south (green dashed line in the middle panel of figure 2) occur during July–September. The burning period corresponds to the tropospheric ozone level for the north but not for the southern part of Thailand. Figure 3 shows the correlation of the tropospheric ozone concentration and the fire occurrence rate. Strong correlations were observed for the northern (0.54) and middle (0.58) parts of Thailand, weak correlations for the north-east (0.34) and negative correlations for the southern part (−0.26) of Thailand.

| Regions   | Trend ± σ (DU/year) | p-value | t | Mean (DU) | Max (DU) |
|-----------|---------------------|---------|---|-----------|---------|
| North     | −0.109 ± 0.4592     | 0.5824  | 0.5808 | 33.7704   | 43.38   |
| Northeast | −0.255 ± 0.7129     | 0.4143  | 0.8768 | 32.9825   | 39.81   |
| Middle    | −0.131 ± 0.7713     | 0.6918  | 0.4160 | 32.5312   | 39.08   |
| South     | −0.303 ± 0.9826     | 0.4793  | 0.7540 | 26.2445   | 32.75   |

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Table 1. The 95% confidence interval for tropospheric ozone trend during the burning season (February–April of eight year analysis).
Including the surrounding regions of Thailand, the correlations became higher for the northern and middle parts (0.77 and 0.51, respectively) and less negative for the southern part (−0.09).

These results suggest that the tropospheric ozone concentrations during burning season originate from fires and were subsequently mixed with anthropogenic pollution other than biomass burning as explained in Amnuaylojaroen et al. (2014). Seasonality and air mass transport is the main reason for these relationships. This would be discussed further in the next section.

The interannual variability of the tropospheric ozone column in Thailand is modulated by the ENSO with the tropospheric ozone column concentrations leading approximately three (3) months in advance of the SST anomalies (determined by the ONI). This lag of SST is due to the coupling characteristic of the atmosphere-ocean system. The 3-month lag of SST anomalies was determined by 1-month lagging and advancing the ONI then calculating the correlation coefficient between the tropospheric ozone column and ONI as shown in figure 4 and table 2.

By subdividing the correlations in figure 4 one for negative ONI values (La Niña) and another for positive ONI values (El Niño), a clearer relationship can be seen as shown in figure 5. The tropospheric ozone column concentration decreases from neutral periods to strong La Niña periods due to enhanced convection diluting the ozone concentration in the tropospheric column. At the same time, increased precipitation during La Niña decreases the chances of the occurrence of fires. Competing with these processes, enhanced convection may produce more thunderstorms wherein lightning NOx (a precursor of tropospheric ozone) can be produced. From neutral periods to strong El Niño periods, the tropospheric ozone column amount also decreases. This is due to suppressed convection inhibiting the production of lightning NOx. The delicate balance of these competing factors causes interannual variability in the tropospheric ozone column (Doherty et al. 2006). Determining the contribution of each of these factors is beyond the scope of this paper.

### Table 2. Correlation coefficients as a function of 1-month ONI shifts for the different regions of Thailand.

| Shift (months) | North | North East | Middle | South |
|---------------|-------|------------|--------|-------|
| −6            | 0.0956| 0.1109     | 0.0842 | 0.1300|
| −5            | 0.0856| 0.0875     | 0.0676 | 0.1131|
| −4            | 0.0094| 0.0595     | 0.0406 | 0.1229|
| −3            | −0.0370| 0.0391     | 0.0182 | 0.1207|
| −2            | −0.0496| 0.0498     | 0.0195 | 0.1432|
| −1            | −0.0107| 0.0966     | 0.0552 | 0.1716|
| 0             | 0.0684| 0.1636     | 0.1144 | 0.2098|
| 1             | 0.1456| 0.2141     | 0.1682 | 0.2294|
| 2             | 0.1860| 0.2311     | 0.1983 | 0.2442|
| 3             | **0.2027**| **0.2338**| 0.2234 | **0.2527**|
| 4             | 0.1943| 0.2271     | **0.2376**| 0.2490|
| 5             | 0.1654| 0.1990     | 0.2243 | 0.2159|
| 6             | 0.1028| 0.1372     | 0.1670 | 0.1370|
| Max           | **0.2027**| **0.2338**| **0.2376**| **0.2527**|

### 3.2. Tropospheric ozone sources and transport in Asia

Most fire occurrences in the northern regions of southeast Asia (i.e. Thailand, Laos, Myanmar and India) occur in April. This is also true for the northern part of Thailand and for the surrounding regions. This leads to elevated tropospheric ozone levels during this period. For the southern part of Thailand and its surrounding regions, most of the fire occur in August and has a small influence to the tropospheric ozone column concentration in this region. The two months of April and August were selected to represent the transport of ozone precursors from the burning region. Since the tropospheric ozone column level in...
April 2006 was the lowest among the other years, and typically high in 2007, 2009 and 2012, these four years were chosen to represent the dynamic transport and the ozone sources.

### 3.2.1. Transport in the northern part of Thailand

Figure 6 shows the locations and the radiative power of the fire occurrences, the fire count density, the distribution of the tropospheric ozone column concentrations and the forward trajectories for April 2006, 2007, 2009 and 2012. A large gradient of approximately 10 DU was observed at 10°N. This can be attributed to the location of the ITCZ, hence the ‘chemical equator’, during this period (Hamilton et al. 2008).

It can also be observed that a substantial number of fires occurred to the north of Thailand especially...
near the borders with Myanmar and northern Laos. A relatively low number of fire occurrences can also be seen in northeast India and southeast China but relatively high tropospheric ozone levels was observed. Sahu et al. 2013 and Kulkarni et al. 2010 reported the ozone over India. Xu and Lin (2011) and Zhang et al. (2014) showed the trends of ozone over China. The ozone levels over these regions were mostly generated from the growing population, expanding industrial sectors and increased anthropogenic activities. However, few fire occurrences were found in northern Philippines, Malaysia, Indonesia and south of China. From the spatial distributions of the fire radiative power and the fire count density, three main areas of burning can be inferred—northeastern India, northern Myanmar and northern Laos. These three main areas of burning were then used as the initial locations for the forward trajectories.

The forward trajectory model outputs show that ozone precursor-rich air masses are generally advected towards the east and northeast in April. These air masses are then blocked by the mountains in the Himalayas, Bhutan and the Chin Hills allowing tropospheric ozone to form and settle along Nepal, north eastern India, Bangladesh and north western Myanmar. Li et al. (2005) and Kar et al. (2010) reported that pollution can be trapped in these regions from north east India and Lin et al. (2009) discussed that the peak plume rise height is in the range of few hundred meters near 1000 m and additional forcing is needed to lift the biomass burning product to cross the height of 3 km topographic elevation. Ozone levels reach about 45 DU in northwestern India and Bangladesh and spread southward to reach the northern part of Thailand and the Bay of Bangal.

In April 2006, the transport of ozone precursor-rich air masses from biomass burning in northern Myanmar advected in a southwestward manner producing seasonally normal ozone levels in the northern part of Thailand and its surrounding regions. In April 2007, 2009 and 2012, the air masses were advected with a wider angle contributing to the ozone passing through northern Thailand and southern of China. Details of the pollution transport to the Pacific Rim have been discussed in Liu et al. 2002.

3.2.2. Transport in the southern part of Thailand

Tropospheric ozone in southern Thailand also shows seasonal variability, as shown in figure 2. The air pollution from biomass burning in the northern part was advected towards the southern part of Thailand during the dry season. During the wet season (June–October) in the northern part of Thailand, the southern part including the surrounding regions become dry due to the influence of the Australian continental air mass. As a result, biomass burning often occurs in Indonesia. However, during this period, the burning season from Indonesia do not have a significant impact on the tropospheric ozone column levels in Thailand and its surrounding regions. Figure 7 shows similar graphs as figure 6 but for August. The 10 DU gradient in the tropospheric ozone level has now migrated upward at approximately 20°N as a result of the migration of the ITCZ and the ‘chemical equator’ (Hamilton et al. 2008).
The El Niño periods of 2006–2007 and 2009–2010 triggered periods of intense biomass burning in the areas of Sumatra and Borneo in Indonesia as shown in figures 2 and 7. However, enhancements in tropospheric ozone column concentration, particularly in the southern portion of Thailand, were not observed. This is due to the northeasterly wind directions generally blowing during this season. This northeasterly wind transports the tropospheric ozone precursors to the ocean as also reported by Srivastava and Sheel (2013) and Sahu et al. (2013). The marine air mass plays an important role in vertical transport. It provides as a sink of tropospheric ozone in which the O(1D) from photolysis of ozone can react with water vapor to produce hydroxyl radicals to reduce the ozone as explained by e.g. Crutzen (1988) and Johnson et al. (1990). Additionally, chlorine and bromine from sea salt can also be another mechanism to destroy tropospheric ozone as explained in Hauglustaine et al. (1999) and Ali et al. (2009). This differs from tropospheric ozone rich air that is transported from the northern region, where there is almost always significant ozone production advected to other regions.

4. Conclusion and recommendations

Tropospheric ozone column concentrations over Thailand, having a background level of about 20 DU, show a seasonal and an interannual variation. The highest tropospheric ozone levels were observed in the northern part of Thailand during the forest-clearing and agricultural burning season that happens from February to April. The mean ozone level during this period is about 33 DU. This is due to the emission of ozone precursor-rich air in this region, which is enhanced by the southwestward transport from northern Myanmar. A slightly non-significant increasing 8-year trend in the tropospheric ozone column was also observed in northern Thailand.

The middle and northeastern parts of Thailand were only nominally affected from the airmass transport from the surrounding regions. Low seasonal variation in the tropospheric ozone column was observed in the southern part of Thailand even though high fire occurrence rates were observed in the southern surrounding regions. This is due to northeasterly advection away from Thailand to the ocean wherein a sink mechanism takes place over the maritime areas.

Interannual variability in the tropospheric ozone column is modulated by ENSO with the concentrations leading SST anomalies by 3 months. The tropospheric ozone column concentrations decrease during both the neutral to strong La Niña and the neutral to the strong El Niño periods. This is due to the low fire occurrence rates occurring during La Niña and less production of lightning NO, during El Niño. El Niño onsets also generally occur during the low biomass burning season. In this study, only a qualitative approach was taken for the transport processes. Quantification of these mechanisms including chemical reactions still has to be performed.

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